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AKERS, Douglas W.

Art Unit

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Examiner Name

Palabrica, R.J.

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In Re Application of:	)	
	)	
AKERS, Douglas, W.	)	Examiner: Palabrica, R.J.
	)	
Serial No. 10/788,743	)	
	)	Group Art Unit: 3663
Filing Date: February 25, 2004	)	
	)	
For: METHOD FOR ON-LINE	)	Confirmation No.: 6111
EVALUATION OF MATERIALS	)	
USING PROMPT GAMMA RAY	)	
ANALYSIS	)	
	)	
Atty Dkt: B-200	)	

**APPEAL BRIEF**

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## TABLE OF CONTENTS

REAL PARTY-IN-INTEREST .....	- 2 -
RELATED APPEALS AND INTERFERENCES.....	- 3 -
STATUS OF THE CLAIMS .....	- 4 -
STATUS OF AMENDMENTS .....	- 5 -
SUMMARY OF CLAIMED SUBJECT MATTER.....	- 6 -
GROUND OF REJECTION TO BE REVIEWED ON APPEAL.....	- 12 -
ARGUMENT .....	- 13 -
Opening Statement .....	- 13 -
I. Whether claims 1-10, 12-19 and 21-23 are patentable under 35 U.S.C. §112, first paragraph, as complying with the enablement requirement.....	- 14 -
II. Whether claims 12-19 and 21-23 are unpatentable under 35 U.S.C. §112, first paragraph, as not enabled for failing to include critical elements.....	- 27 -
III. Whether claims 1-10, 12-19 and 21-23 are patentable under 35 U.S.C. §112, second paragraph, as distinctly claiming the subject matter of the invention.....	- 31 -
IV. Whether claims 12-19 and 21-23 are unpatentable under 35 U.S.C. §112, second paragraph, for failing to include critical elements.....	- 37 -
CONCLUSION .....	- 40 -
CLAIMS APPENDIX .....	- 42 -
EVIDENCE APPENDIX.....	- 47 -
RELATED PROCEEDINGS APPENDIX.....	- 48 -
REFERENCES APPENDIX.....	- 49 -

## TABLE OF AUTHORITIES

Cases:	Page
<i>CFMT, Inc. v. Yieldup Int’l Corp.</i> , 349 F.3d 1333, 1338 (Fed. Cir. 2003) ....	15 -, 17 -, 25 -
<i>Credle v. Bond</i> , 25 F.3d 1566, 1576 (Fed. Cir. 1994).....	30 -
<i>Ex parte Akers</i> , Appeal No. 2005-0855, at 8 (Bd. Pat. App. & Interf. 2005).-	12 -, 34 -, 47 -
<i>Ex parte Kato, et al.</i> , 2002 WL 465390 (Bd. Pat. App. & Interf. 2002).....	27 -
<i>Ex parte Kopetzki</i> , 2001 WL 11197754 (Bd. Pat. App. & Interf. 2001).....	14 -, 21 -, 25 -
<i>Ex parte Lemelson</i> , 2002 WL 32334419, *3 (Bd. Pat. App. & Interf. 2002).....	<i>passim</i>
<i>Ex parte Nakano, et al.</i> , 1998 WL 1735647 (Bd. Pat. App. & Interf. 1998).....	27 -
<i>Ex parte Noguchi</i> , 2002 WL 1801470 (Bd. Pat. App. & Interf. 2002) .....	14 -, 21 -
<i>Ex parte Trummer</i> , 1997 WL 33106615, *4 (Bd. Pat. App. & Interf. 1997) .....	37 -, 39 -
<i>Ex parte Voordum</i> , 2002 WL 1801166, *4 (Bd. Pat. App. & Interf. 2002) .....	36 -
<i>Gould v. Missinghoff</i> , 229 USPQ 1 (D.D.C. 1985).....	15 -
<i>In re Angstadt</i> , 537 F.2d 498, 190 USPQ 214 (CCPA 1976).....	14 -
<i>In re Brebner</i> , 455 F.2d 1402, 173 USPQ 169 (CCPA 1972) .....	15 -
<i>In re Buchner</i> , 929 F.2d 660, 18 USPQ2d 1331 (Fed. Cir. 1991) .....	15 -
<i>In re Curtis</i> , 354 F.3d 1347, 1357 (Fed. Cir. 2004) .....	31 -
<i>In re Goffe</i> , 542 F.2d 564, 567, 191 USPQ 429, 431 (CCPA 1976).....	26 -
<i>In re Hyatt</i> , 708 F.2d 712, 715 (Fed. Cir. 1983) .....	31 -, 33 -
<i>In re Johnson</i> , 558 F.2d 1008, 1015, 194 USPQ 187 (CCPA 1977) .....	31 -
<i>In re Marzocchi</i> , 439 F.2d 220, 169 USPQ 367 (CCPA 1979) .....	15 -
<i>In re Mayhew</i> , 527 F.2d 1229, 1233 (C.C.P.A. 1976) .....	26 -
<u><i>In re Miller</i></u> , 441 F.2d 689, 169 USPQ 597, 600 (CCPA 1971).....	13 -, 34 -

*In re Oetiker*, 977 F.2d 1443, 1445 (Fed. Cir. 1992) ..... - 31 -, - 32 -

*In re Wands*, 858 F.2d 731, 8 USPQ2d 1400 (Fed. Cir. 1988).....*passim*

*Miles Laboratories, Inc., v. Shandon, Inc.*, 27 USPQ2d 1123 (Fed. Cir. 1993)..... - 30 -

**Statutes and Regulations:**

35 U.S.C. § 112, first paragraph.....*passim*

35 U.S.C. § 112, second paragraph.....*passim*

MPEP 2164.01(a)..... - 14 -

MPEP 2164.08 ..... - 26 -, - 27 -

MPEP 2172.01 ..... - 37 -

MPEP 2173.02 ..... - 30 -, - 31 -, - 33 -



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
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USING PROMPT GAMMA RAY	)	
ANALYSIS	)	
	)	
Atty Dkt: B-200	)	

**APPEAL BRIEF**

To: Commissioner of Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

This Appeal Brief is submitted in response to the non-final rejections of the claims in the office action, mailed January 16, 2007. A notice of appeal was filed on January 26, 2007.

**REAL PARTY-IN-INTEREST**

The assignee of the entire right, title, and interest in the patent application is Bechtel  
BWXT Idaho, LLC.

### **RELATED APPEALS AND INTERFERENCES**

There are three related appeals of other United States patent applications that may directly affect, or be directly affected by, or have a bearing on, the Board's decision. Divisional application, serial no. 10/269,807, filed October 10, 2003, came before the Board in Appeal No. 2005-0855. The Board mailed its decision to the appellant on May 31, 2005. United States Patent No. 7,058,153 subsequently issued on June 6, 2006. Divisional application, serial no. 10/383,096, filed March 5, 2003, is currently pending on appeal in the briefing stage. In parent application, serial no. 09/932,531, filed August 17, 2001, an appeal brief was filed on June 21, 2006. Thereafter, on February 23, 2007, a notice of allowance was issued, allowing the claims at issue in the appeal. The current application is a continuation-in-part of parent application, serial no. 09/932,531, and a continuation-in-part of divisional application, serial no. 10/383,096.

There are currently no related interferences known to appellant, appellant's legal representative, or the assignee which will directly affect, or be directly affected by, or have a bearing on, the Board's decision.



### **STATUS OF THE CLAIMS**

Claims 1-10, 12-19 and 21-23 are pending in the application. Claims 11, 20 and 24 are cancelled. Claims 1-10, 12-19 and 21-23 currently stand rejected. The rejections of claims 1-10, 12-19 and 21-23 are appealed.

In the present application, the examiner provisionally rejected claim 2 under the judicially-created doctrine of double patenting over claim 3 of co-pending divisional application, serial no. 10/383,096. The appellant did not traverse this provisional rejection and has agreed to file the appropriate terminal disclaimer in the appropriate application at the appropriate time (i.e., upon the indication of allowance of either claim 2 in this application or claim 3 of the co-pending application).

### **STATUS OF AMENDMENTS**

No amendments were filed or entered subsequent to the non-final office action mailed on January 16, 2007.

## **SUMMARY OF CLAIMED SUBJECT MATTER**

The present invention is directed to a method for evaluating a material specimen by bombarding it with neutrons to create prompt gamma rays. The invention as claimed is summarized below with reference to the independent claims and claims separately argued. Claims 1 and 12 are independent claims. Claims 2, 13-19, 21 and 23 are dependent claims argued separately. The claims contain reference numerals and reference to the specification and drawings. All references are shown in the application at least where indicated herein.

1. A method (226; Fig. 9; ¶72) for evaluating a material specimen (12, 112, 212; Figs. 1, 7-9; ¶¶23-31, 33-34, 37-40, 43, 47, 50-53, 055-57, 59-63, 66, 68-69, 71-75, 77), comprising:

mounting (228, Fig. 9; ¶¶37, 59-61, 72-74, 77) a neutron source (14, 54, 114, 214, 254; Figs. 1, 7-9; ¶¶23-30, 32-34, 36-37, 50, 52, 56-63, 72-74, 77) adjacent the material specimen (12, 112, 212);

mounting (Fig. 9; ¶¶38, 40, 66-69) a detector (16, 30, 32, 116, 130, 216, 230; Figs. 1, 4, 6-9; ¶¶23-24, 36, 38-42, 44-46, 49, 52-54, 56, 60, 65-72, 74-77) adjacent the material specimen (12, 112, 212);

bombarding (232; Fig. 9; ¶¶23-24, 30-31, 34, 37, 50, 56, 59-61, 73) the material specimen (12, 112, 212) with neutrons (Figs. 1, 7-9; ¶¶30-34, 37, 50, 52, 56-63, 73) from the neutron source (14, 114, 214) to create prompt gamma rays (Figs. 1, 7-9; ¶¶23-24, 27, 30, 34, 37-39, 41, 43-45, 49-53, 57, 60-61, 63, 66-67) within the material specimen (12, 112, 212), some of the prompt gamma rays being emitted from the material specimen (12, 112, 212), some of the prompt gamma rays resulting in the formation of positrons (Figs. 1, 7-9; ¶¶23-25, 27, 30-31, 34, 37, 43, 50-52, 56-57, 60-61, 63) within the material specimen (12, 112, 212) by pair production (18, 118, 218; Figs. 1, 7-9; ¶¶23, 28, 30-31, 37, 50, 52, 57, 60-61, 63);

collecting (70, 82, 234; Figs. 3, 6, 9; ¶75) positron annihilation data (22, 122, 222; Figs. 1, 3, 4, 6-9; ¶¶ 38, 42-45, 49, 53-54, 67, 70-71, 75-77) by detecting with the detector (16, 30, 32, 116, 130, 216, 230) a plurality of emitted annihilation gamma rays (Figs. 1, 7-9; ¶¶23-24, 30, 38-39, 41, 43, 45, 51-53, 57, 66-67) resulting from the annihilation (¶¶24, 30, 43-44, 57) of positrons, the detector (16, 30, 32, 116, 130, 216, 230) producing the positron annihilation data (22, 122, 222);

processing (84, 236; Figs. 6, 9; ¶¶26, 42, 47, 49, 53-54, 67, 70, 75, 77) collected positron annihilation data (22, 122, 222) in accordance with a Doppler-broadening algorithm (40; Figs. 2, 6; ¶¶26-27, 38, 42, 47-49, 51, 70, 75); and

continuing (238; Fig. 9; ¶75-76) to collect and process positron annihilation data (22, 122, 222) to measure an accumulation of lattice damage (¶¶24-26, 43, 47, 60-61, 67-69, 71, 74, 77) over time.

2. The method of claim 1, further comprising:

collecting (234; Fig. 9, ¶75-76) prompt gamma ray data (20, 120, 220; Figs. 1, 4, 7-9, ¶¶24-25, 38, 42-45, 49, 53-54, 67, 70-71, 75-77) by detecting with the detector (16, 30, 32, 116, 130, 216, 230) a plurality of emitted prompt gamma rays (Figs. 1, 7-9; ¶¶23-24, 27, 30, 34, 37-39, 41, 43-45, 49-53, 57, 60-61, 63, 66-67), the detector (16, 30, 32, 116, 130, 216, 230) producing the prompt gamma ray data (20, 120, 220);

calculating (72; Fig. 3; ¶¶25, 42-46, 51, 70) positron lifetime data (¶¶25, 27, 42-46, 51, 53, 70) from the positron annihilation data (22, 122, 222) and the prompt gamma ray data (20, 120, 220); and

continuing (238; Fig. 9; ¶¶75-76) to collect positron annihilation data (22, 122, 222) and prompt gamma ray data (20, 120, 220) and calculate (72) positron lifetime data to measure an accumulation of lattice damage over time.

12. A method for evaluating a material specimen (12, 112, 212; Figs. 1, 7-9; ¶¶23-31, 33-34, 37-40, 43, 47, 50-53, 055-57, 59-63, 66, 68-69, 71-75, 77), comprising:

mounting (228, Fig. 9; ¶¶37, 59-61, 72-74, 77) a neutron source (14, 54, 114, 214, 254; Figs. 1, 7-9; ¶¶23-30, 32-34, 36-37, 50, 52, 56-63, 72-74, 77) adjacent the material specimen (12, 112, 212);

mounting (Fig. 9; ¶¶38, 40, 66-69) a detector (16, 30, 32, 116, 130, 216, 230; Figs. 1, 4, 6-9; ¶¶23-24, 36, 38-42, 44-46, 49, 52-54, 56, 60, 65-72, 74-77) adjacent the material specimen (12, 112, 212);

bombarding (232; Fig. 9; ¶¶23-24, 30-31, 34, 37, 50, 56, 59-61, 73) the material specimen (12, 112, 212) with neutrons (Figs. 1, 7-9; ¶¶30-34, 37, 50, 52, 56-63, 73) from the neutron source (14, 54, 114, 214, 254) to create prompt gamma rays (Figs. 1, 7-9; ¶¶23-24, 27, 30, 34, 37-39, 41, 43-45, 49-53, 57, 60-61, 63, 66-67) within the material specimen (12, 112, 212), some of the prompt gamma rays being emitted from the material specimen (12, 112, 212), some of the prompt gamma rays resulting in the formation of positrons (Figs. 1, 7-9; ¶¶23-25, 27, 30-31, 34, 37, 43, 50-52, 56-57, 60-61, 63) within the material specimen (12, 112, 212) by pair production (18, 118, 218; Figs. 1, 7-9; ¶¶23, 28, 30-31, 37, 50, 52, 57, 60-61, 63);

collecting (70, 82, 234; Figs. 3, 6, 9; ¶75) positron annihilation data (22, 122, 222; Figs. 1, 3, 4, 6-9; ¶¶38, 42-45, 49, 53-54, 67, 70-71, 75-77) by detecting with the detector

(16; 30, 32, 116, 130, 216, 230) a plurality of emitted annihilation gamma rays (Figs. 1, 7-9); ¶¶23-24, 30, 38-39, 41, 43, 45, 51-53, 57, 66-67) resulting from the annihilation (¶¶24, 30, 43-44, 57) of positrons, the detector (16, 30, 32, 116, 130, 216, 230) producing the positron annihilation data (22, 122, 222);

storing (¶¶ 71, 76) the positron annihilation data (22, 122, 222) on a data storage system (225; Fig. 8; ¶¶ 71, 76) for later retrieval and processing; and

continuing to collect and store positron annihilation data (22, 122, 222), the continued collected and stored positron annihilation data (22, 122, 222) being indicative of an accumulation of lattice damage (¶¶24-26, 43, 47, 60-61, 67-69, 71, 74, 77) over time.

13. The method of claim 12, further comprising:

collecting prompt gamma ray data (20, 120, 220) by detecting with the detector (16, 30, 32, 116, 130, 216, 230) a plurality of emitted prompt gamma rays, the detector (16, 30, 32, 116, 130, 216, 230) producing the prompt gamma ray data (20, 120, 220) ;

storing prompt gamma ray data (20, 120, 220) on the data storage system (225) for later retrieval and processing; and

continuing to collect and store prompt gamma ray data (20, 120, 220) , the continued collected and stored prompt gamma ray data (20, 120, 220) being indicative of an accumulation of lattice damage over time.

14. The method of claim 12, wherein said mounting (228) a neutron source (14, 54, 114, 214, 254) adjacent the material specimen (12, 112, 212) comprises mounting (228) the neutron source (14, 54, 114, 214, 254) to the material specimen (12, 112, 212).

15. The method of claim 14, wherein said mounting a detector (16, 30, 32, 116, 130, 216, 230) adjacent the material specimen (12, 112, 212) comprises mounting the detector (16, 30, 32, 116, 130, 216, 230) to the material specimen (12, 112, 212).
16. The method of claim 15, further comprising positioning a shield (56, 256; Figs. 1, 8; ¶¶33, 62) adjacent the neutron source (14, 54, 114, 214, 254) to absorb stray neutrons.
17. The method of claim 16, further comprising positioning a moderator (60, 260; Figs. 1, 8; ¶¶34-36, 63-65) between the neutron source (14, 54, 114, 214, 254) and the material specimen (12, 112, 212).
18. The method of claim 12, wherein mounting (228) a neutron source (14, 54, 114, 214, 254) adjacent the material specimen (12, 112, 212) comprises mounting (228) an isotopic neutron source (54, 254) adjacent the material specimen (12, 112, 212).
19. The method of claim 12, wherein continuing to collect and store positron annihilation data (22, 122, 222) is performed while the material specimen (12, 112, 212) is in service.
21. The method of claim 12, further comprising:  
retrieving stored positron annihilation data (22, 122, 222); and  
processing (84, 236; Figs. 6, 9; ¶¶26, 42, 47, 49, 53-54, 67, 70, 75, 77) the positron annihilation data (22, 122, 222) in accordance with a Doppler-broadening algorithm (40; Figs.

2, 6; ¶¶26-27, 38, 42, 47-49, 51, 70, 75) to produce output data indicative of an accumulation of lattice damage over time.

23. The method of claim 12, further comprising removing the neutron source (14, 54, 114, 214, 254) before collecting positron annihilation data (22, 122, 222).



**GROUND OF REJECTION TO BE REVIEWED ON APPEAL**

1. Whether claims 1-10, 12-19 and 21-23 are patentable under 35 U.S.C. §112, first paragraph, as complying with the enablement requirement.
2. Whether claims 12-19 and 21-23 are unpatentable under 35 U.S.C. §112, first paragraph, as not enabled for failing to include critical elements.
3. Whether claims 1-10, 12-19 and 21-23 are patentable under 35 U.S.C. §112, second paragraph, as distinctly claiming the subject matter of the invention.
4. Whether claims 12-19 and 21-23 are unpatentable under 35 U.S.C. §112, second paragraph, for failing to include critical elements.

## ARGUMENT

### Opening Statement

The Board will undoubtedly experience a profound sense of déjà-vu in this appeal. It once again raises many of the same issues that the Board has already decided, yet must decide again here (as well as, in the appeal of application serial no. 10/383,096). Once again, the examiner has rejected all the claims in this application for indefiniteness and lack of enablement. Yet, in his quest to brand all the claims indefinite and not enabled, he has based his rejections primarily on limitations that are not part of the claims at issue in *this* application. However, even where the examiner's rejections were actually tied to claim limitations requiring use of a Doppler-broadening algorithm, for example, those claims are enabled and sufficiently definite. The Board reached the same conclusion in a related case, reversing this examiner's rejections of lack of enablement and indefiniteness. The Board's words apply equally well here:

[W]hile the appellant's disclosure does not convey much detail as to the various algorithms described therein, these algorithms admittedly are generally known in the art. The examiner has failed to advance any cogent reasoning as to why the disclosure would not have enabled a person of ordinary skill in the art to employ these algorithms without undue experimentation.

*Ex parte Akers*, Appeal No. 2005-0855, at 8 (Bd. Pat. App. & Interf. 2005) (slip op.). The Board also reversed the examiner's rejections based on indefiniteness of the claims as follows:

The examiner views the appealed claims to be indefinite for reasons essentially similar to those listed above in connection with the enablement rejection. In this regard, the examiner points to the claim limitations relation to . . . the positron lifetime, Doppler-

broadening, three-dimensional imaging and activation/analysis process algorithms. . . . [T]he examiner's criticism of the claim limitations pertaining to the algorithms . . . focus[es] on the breadth of these limitations. It is well settled, however, that mere breadth

does not equate to indefiniteness. In re Miller, 441 F.2d 689, 169 USPQ 597, 600 (CCPA 1971).

*Id.* at 9.

The examiner's new grounds of rejection do not demonstrate that the claims are unpatentable either. The examiner argues that the claims are not enabled and incomplete because they fail to include a "critical" processing step. No part of the specification describes a processing step as "key" or "critical" or "essential." The fact that the specification discloses a step of processing does not render that step essential. Moreover, in making these rejections, the examiner has sidestepped portions of the specification that describe embodiments for collecting and storing data without processing, thus demonstrating that processing is not essential.

Although steadfast in his arguments, the examiner's rejections lack what is required to sustain them -- evidence from the specification, including the abstract and the claims originally filed, to show that any of these claims are indefinite or not enabled. Thus, the examiner's rejections must be reversed.

**I. Whether claims 1-10, 12-19 and 21-23 are patentable under 35 U.S.C. §112, first paragraph, as complying with the enablement requirement.**

**A. Legal Standard For Rejecting Claims Under 35 U.S.C. §112, First Paragraph**

The legal standard for determining whether the disclosure provides a sufficient description of the invention is whether a person reasonably skilled in the art could make or use the invention without undue experimentation based on the disclosure and on information known in the art. *United States v. Telectronics, Inc.*, 857 F.2d 778, 8 USPQ2d 1217 (Fed. Cir. 1988). The fact that experimentation may be complex does not necessarily make it undue if

the art typically engages in such experimentation. *In re Wands*, 858 F.2d 731, 8 USPQ2d 1400 (Fed. Cir. 1988). See *Ex parte Lemelson*, 2002 WL 32334419, \*3 (Bd. Pat. App. & Interf. 2002) (“The fact that an invention is sophisticated and complex does not, by itself, lead to the conclusion that undue experimentation would be required to make and use the invention.”) That is, the test of enablement is not whether any experimentation is required, but whether, if experimentation is necessary, it is undue. *In re Angstadt*, 537 F.2d 498, 190 USPQ 214 (CCPA 1976).

The factors to be considered when determining whether a claim limitation is enabled and whether any necessary experimentation is “undue” include, but are not limited to: (1) the breadth of the claims; (2) the nature of the invention; (3) the state of the prior art; (4) the level of one of ordinary skill; (5) the level of predictability in the art; (6) the amount of direction provided by the inventor; (7) the existence of working examples; and (8) the quantity of experimentation needed to make or use the invention based on the content of the disclosure. *In re Wands*, 858 F.2d at 737; MPEP 2164.01(a). It is improper to conclude that a disclosure is not enabling based on an analysis of only one of the above factors while ignoring one or more of the others. See, e.g., *Ex parte Noguchi*, 2002 WL 1801470 (Bd. Pat. App. & Interf. 2002) (supplying information directed to one factor is insufficient); *Ex parte Kopetzki*, 2001 WL 11197754 (Bd. Pat. App. & Interf. 2001) (merely reciting facts without reference to the factors is insufficient); MPEP 2164.01(a). Moreover, whether the specification is enabling must be determined with reference only to the limitations claimed. *CFMT, Inc. v. Yieldup Int’l Corp.*, 349 F.3d 1333, 1338 (Fed. Cir. 2003).

With regard to the burden of proof required to support a rejection under §112, the Patent Office is required to assume that the specification complies with the enablement

provision of §112 unless it has acceptable evidence or reasoning to suggest otherwise. *See, e.g., In re Marzocchi*, 439 F.2d 220, 169 USPQ 367 (CCPA 1979). The Patent Office thus must provide reasons, supported by the record as a whole, why the specification is not enabling. Then and only then does the burden shift to the applicant to show that one of ordinary skill in the art could have practiced the claimed invention without undue experimentation. *Gould v. Missinghoff*, 229 USPQ 1 (D.D.C. 1985), *aff'd in part, vacated in part, and remanded sub. nom., Gould v. Quigg*, 822 F.2d 1074, 3 USPQ2d 1302 (Fed. Cir. 1987). Mere conclusionary statements as to the level of ordinary skill in the art are not a sufficient basis for a rejection under 35 U.S.C. §112. *In re Brebner*, 455 F.2d 1402, 173 USPQ 169 (CCPA 1972). In addition, the law does not require, and indeed prefers, that a patent specification omit that which is well-known. *In re Buchner*, 929 F.2d 660, 18 USPQ2d 1331 (Fed. Cir. 1991).

B. Response to Examiner's Rejections

The examiner failed to carry his prima facie burden of demonstrating that claims 1-10, 12-19 and 21-23 are not enabled as required by §112, first paragraph. First, the examiner's rejections are based on limitations that are not part of the claims. Second, where the examiner did base his rejections on at least one limitation actually contained in the claims, the examiner failed to analyze the enablement issue with reference to the eight required factors. Finally, given that Doppler-broadening (and other) algorithms are well-known in the art, the level of detail contained in the specification sufficiently enables the claims.

1. Rejections are improperly based on limitations not in the claims.

As an initial matter, the examiner argues that “the claims require the use of algorithms (i.e., Doppler-broadening and positron lifetime), detector(s), a neutron source and a data

processor. Thus to enable an artisan to make and use an operative embodiment, one must first know which particular algorithm(s) to use, and how and in what manner the data from the detectors are to be analyzed in the 'data processor'." Office Action (1/16/07), pp. 6-7; Final Office Action (3/1/06), p. 2. A fundamental deficiency in the examiner's argument is that none of the claims requires use of a data processor. And, none of the claims requires use of a positron lifetime algorithm. Since the examiner's rejections are based on limitations not found in the claims, they should be reversed.

a. Independent claim 12 and dependent claims 13-19 and 23

Independent claim 12 and dependent claims 13-19 and 23 require neither processing in accordance with an algorithm nor use of a data processor. Therefore, the examiner's §112 rejections of these claims for lack of enablement of those limitations are improper on their face and should be reversed on that ground alone.

b. Independent claim 1 and dependent claims 2 and 21

Now the appellant turns to independent claim 1 and dependent claim 21, both of which contain the limitation, "processing . . . in accordance with a Doppler-broadening algorithm." Claim 2 claims the method of claim 1 further comprising "calculating positron lifetime data;" however, claim 2 does not require use of a positron lifetime algorithm. The examiner argues that the claims are not enabled because undue experimentation is necessary to select algorithms (including Doppler-broadening, positron lifetime, 3D imaging and selective activation algorithms) that will (1) obtain accurate detection of lattice defects (i.e., not too few and not too many); (2) obtain reasonable statistical analyses; (3) determine which algorithm (in conjunction with selection of "a suitable data processor") "actually provides a reasonable determination of the presence of enough lattice defects to be of concern;" and (4) determine

“whether there are present enough lattice defects to cause metal fatigue, etc. . . .” Office Action (1/16/07), p. 10; Final Office Action (3/1/06), p. 6. The examiner further argues that the appellant has “refused to disclose how these algorithms have to be combined,” referring to Doppler-broadening and positron lifetime algorithms. Office Action (1/16/07), p. 11; Final Office Action (3/1/06), p. 7 (underlining omitted). However, these limitations are not in the claims and, therefore cannot be considered when determining whether the specification enables the claims.

None of these claims requires a data processor; positron lifetime, 3-D imaging or selective activation algorithms; accuracy; reasonable statistical analysis; “provid[ing] a reasonable determination of the presence of enough lattice defects to be of concern;” or determining “whether there are present enough lattice defects to cause metal fatigue.” Office Action (1/16/07), p. 10; Final Office Action (3/1/06), p. 6. Similarly, none of the claims requires combining Doppler-broadening and positron lifetime algorithms. As the Federal Circuit has held, “Title 35 does not require that a patent disclosure enable . . . a perfected, commercially viable embodiment absent a claim limitation to that effect.” *CFMT, Inc. v. Yieldup Int’l Corp.*, 349 F.3d 1333, 1338 (Fed. Cir. 2003). Therefore, the examiner cannot evaluate enablement based on limitations not contained in the claims. *Id.*, 349 F.3d at 1338 (since claims contained no standard for cleanliness, claims for cleaning methods were enabled for any level of contaminant removal); *Ex parte Lemelson*, 2002 WL 32334419, \*4.

Ignoring the plain language of the claims, the examiner nonetheless insists that the claims require the use of algorithms, a detector, a neutron source and a data processor. Office Action (1/16/07), p. 16. The examiner makes this statement based on his belief that “Appellant does not disclose any alternative method of performing the material evaluation

that does not require these four elements.” Office Action (1/16/07). However, that is not the case. Paragraph 71 of the specification describes “the third embodiment” wherein “the data processing system serves merely as a data collection device”:

[0071] Alternatively, the third embodiment 210 to be used in a long-term arrangement, such as, for example to measure the build-up or accumulation of lattice defects during the service life (or some portion of the service life) of the material specimen 212, the data processing system 224 could be simplified considerably to where the *data processing system 224 serves merely as a data collection device*, collecting the data (e.g., the prompt gamma ray data 220 and/or the positron annihilation data 222) produced by the detector assembly 216, and storing the data in a data storage system 225 for later retrieval and processing in accordance with the teachings provided herein. . . . Thereafter, the collected and stored data *may be* retrieved (i.e., downloaded) and processed in accordance with the teaching herein in order to provide information indicative of the build-up or accumulation of lattice defects during the collection period.

*See infra* at pp. 29-30 (quoting the abstract which describes a similar embodiment). Contrary to the examiner’s arguments, no processing using an algorithm is disclosed.

With respect to his rejections of claim 2, the examiner argues that a data processor and a positron lifetime algorithm must be included in claim 2 as essential elements; otherwise, claim 2 will not be enabled. Office Action (1/16/07). However, this argument cannot be sustained in light of passages from the specification stating that “the data processing system 224 *may* process the prompt gamma ray data 220 and positron annihilation data 222 in accordance with a positron lifetime algorithm . . .” *See* Specification, ¶ 70 (emphasis added). The examiner argues that because no other method for calculating positron lifetime is disclosed, “there must be other undisclosed means” for calculating positron lifetime, insinuating that the appellant has failed to disclose the best mode (which is denied). First, the disclosure of only one embodiment for making and using the invention satisfies the



enablement requirement. MPEP 2163.01(b). Second, the examiner is not entitled to assume that these allegedly “undisclosed means” – to the extent they exist at all (which is denied) – are better than that disclosed in the specification, for they just as well could be worse. And, finally, the examiner has provided no evidence that the best mode has not been disclosed.

c. Figure 2

Similarly, but without reference to any claims, the examiner objects to the sufficiency of the disclosure in Figure 2, regarding a feedback arrangement between the data processing system 24 and the algorithms 38, 40. Office Action (1/16/07), p. 12; Final Office Action (3/1/06), p. 8. However, the examiner has not tied these arguments to the elements of any claim to show how such elements are not enabled. *CFMT, Inc.*, 349 F.3d at 1338 (level of disclosure necessary for enablement varies with the scope of the claims). Therefore, the examiner’s arguments in this regard cannot be used to demonstrate a prima facie case of non-enablement.

d. *Ex parte Lemelson*

The case of *Ex parte Lemelson*, 2002 WL 32334419, presents similar facts to the ones here and is persuasive in its analysis of enablement. In that case, the invention was directed to a method and apparatus for controlling the travel of a powered vehicle, comprising calculating whether the vehicle and an object are on a collision course by measuring the distance to and relative velocity between the two, and using fuzzy logic interference rules to determine a combination of steering and acceleration to avoid a collision. The examiner rejected the claims for lack of enablement on grounds almost identical to the ones here:

The rejection also states that the disclosure is insufficient for teaching how to distinguish one object from another object or how to determine distance and relative velocity of each object. . . . The examiner observes that the object detection and recognition and the

fuzzy logic rules require complex algorithms and extensive processing that are not sufficiently described in the disclosure [citation omitted]. In the final rejection the examiner added that the complexity of putting all the features into one real-time system required undue experimentation.

*Id.*, at \*2. The appellants argued that fuzzy logic and image analysis were techniques well-known in the art. The Board reversed the examiner's rejections. First, the Board held that the examiner improperly based his rejections on a real-time operation requirement not included in the claims. It also held, "Second, the examiner's findings of undue experimentation are mere conclusions based on the examiner's own speculations. The examiner essentially finds that there would be undue experimentation because the claimed invention requires sophisticated and complex operations. The fact that an invention is sophisticated and complex does not, by itself, lead to the conclusion that undue experimentation would be required to make and use the invention." *Id.* at \*3. The examiner here argues that the instant case differs from the *Lemelson* case because he did show undue experimentation and he did show that the elements associated with the rejection are required (both of which are denied). However, arguing that the result of the case is different does not distinguish *Ex parte Lemelson* from this case on the facts. The Board's analysis in the *Lemelson* case applies equally well here, demonstrating that the examiner here failed to carry his burden.

e. Dependent claims 2-10 and 22

In addition, claims 2-10 and 22 depend from claim 1 and are also allowable at least for the reasons that claim 1 is allowable.

2. The eight required factors were not addressed.

While the examiner sets forth a laundry list of variables that he argues one of ordinary skill in the art must consider when choosing an appropriate Doppler-broadening algorithm,

positron lifetime algorithm, or other algorithm, the examiner has nonetheless failed to meet his prima facie burden. As an initial matter, again, (with the exception of Doppler-broadening) none of the claims at issue here requires processing in accordance with positron lifetime or other algorithms, so those “limitations” may not be considered in determining whether the claims are enabled. In addition, the examiner failed to undertake the required analyses to demonstrate lack of enablement.

Only claims 1 and 21 contain a limitation that the examiner argues was not enabled. That limitation is “processing . . . in accordance with a Doppler-broadening algorithm. As stated above, while claim 2 claims “calculating positron lifetime data,” it does not require use of a positron lifetime algorithm. However, in rejecting claims 1, 2 and 21 (as well as all the other claims) for non-enablement, the examiner was required to analyze each of the eight required factors set forth in *In re Wands*, 858 F.2d 731, 737 (Fed. Cir. 1988), with reference to the claims, which he did not. Nor did the examiner supply facts to support each and every factor required by *In re Wands*. The Board has repeatedly reversed similar rejections, holding that the examiner failed to meet his prima facie burden. See, e.g., *Ex parte Noguchi*, 2002 WL 1801470 (Bd. Pat. App. & Interf. 2002) (supplying information directed to one factor is insufficient); *Ex parte Kopetzki*, 2001 WL 11197754 (Bd. Pat. App. & Interf. 2001) (merely reciting facts without reference to the factors is insufficient). Even assuming that the examiner put forth any “facts” (which is denied), the “mere citing of a series of facts, without relating them to the criteria for determining undue experimentation set forth in *In re Wands*, is not sufficient to establish a prima facie case of nonenablement.” *Ex parte Kopetzki*, 2001 WL 11197754, at \*3. In addition, by reviewing the variables that one of ordinary skill in the art would know to consider, the examiner makes the appellant’s point that one of ordinary skill in

the art would well know how to pick a suitable Doppler-broadening algorithm (or positron lifetime algorithm, assuming one were even required) and select appropriate constants, making any experimentation routine and not undue.

3. The specification is enabling.

Finally, given that Doppler-broadening algorithms are well-known, the level of detail provided in the present specification is sufficiently enabling, as the Board determined with reference to the appeal in the divisional case issued as U.S. Patent No. 7,058,153. *See supra* at p. 12. Moreover, even if a positron lifetime algorithm were required by claim 2 (which is denied), positron lifetime algorithms are also well-known, rendering the level of detail provided in the specification enabling as to that limitation, too.

The appellant specifically states in the specification that the Doppler-broadening algorithms and positron lifetime algorithms are well-known in the art and could be readily implemented by persons having ordinary skill in the art after having become familiar with the teachings of the present invention. Specification, ¶¶ [0043]-[0049]. Because these algorithms are well-known to those of ordinary skill in the art who are familiar with their applications, the specification need not and does not describe them further. It is undisputed that these algorithms are well-known. Office Action (1/16/07), p. 18. Since that fact is undisputed, the examiner was bound to establish that the amount of experimentation in using these algorithms familiar to those of ordinary skill in the art would be undue. As explained above, he failed to make such a showing because he did not analyze the undue experimentation factors from *In re Wands*.

During prosecution, the appellant provided examples of issued patents to show the general level of detail required to enable similar inventions for those of ordinary skill in the

art. *See* Amendment (1/13/06), p. 12. In his Summary of Argument presented in the appellant's Amendment dated January 13, 2006, the appellant stated that "the best measure of level of disclosure required in this particular field of endeavor can be obtained by reviewing issued U.S. patents in the same field." Amendment (1/13/06), p. 7. The examiner argued that U.S. Patent No. 6,178,218 to Akers ("Akers") and U.S. Patent No. 4,064,438 to Alex, et al. ("Alex") cannot be used by analogy to show the appropriate level of detail required for enablement. Office Action (1/16/07), p. 13; Final Office Action (3/1/06), p. 9. However, the examiner's arguments are not well-founded.

Specifically, the examiner argued that Akers cannot be used because his Doppler-broadening algorithm "is not modified by feed back from the data processing system. . . ." Office Action (1/16/07), p. 13; Final Office Action (3/1/06), p. 9. However, the claims at issue in this appeal do not contain as limitations either use of a data processing system or modification of the Doppler-broadening algorithm through feedback. Basing non-enablement rejections on limitations not found in the claims is improper.

Further, the examiner argues that Alex cannot be used to demonstrate enablement by analogy because Alex uses a positron lifetime algorithm for a different purpose than do the claims of the present invention. As an initial matter, none of the claims at issue here requires use of a positron lifetime algorithm. While claim 2 claims "calculating positron lifetime data," it does not require use of a positron lifetime algorithm either. In any event, the examiner misses the point. The point is that the description in Alex enables his claim limitation of a method "wherein the variations in annihilation characteristics within the tested material are detected by positron lifetime measurements." The description of positron lifetime measurements in Alex contains even less detail than the present specification, merely

referring to one page of a previously published book and one issued patent. *Compare* Alex, Col. 6, Ins. 60-66 *with* Specification ¶¶ [0043]-[0046]. If such a brief description in Alex were considered enabling, then the more detailed description of the present specification must also be considered enabling to one of ordinary skill in the art. Finally, the other references previously cited by the examiner (i.e., Derlet, Bandžuch, Zhu, and Shaffer) illustrate that persons having ordinary skill in the art are well-aware of the appropriate use and description of Doppler-broadening and positron lifetime algorithms, as well as the various issues and complexities involved in selecting, modifying and combining any of these algorithms, rendering any experimentation required routine and not undue.

The examiner argues that “the specification indicates that there are different known Doppler-broadening algorithms but, fails to disclose which of these known algorithms would be suitable for use in the present invention.” Office Action (1/16/07), p. 7; Final Office Action (3/1/06), p. 3. That is not the case. The specification specifically discloses a suitable algorithm when it states that “in one preferred embodiment of the invention, the Doppler-broadening algorithm 40 may comprise the Doppler-broadening algorithm described in U.S. Patent No. 6,178,218 B1 . . . .” Specification, ¶[0048]. Contrary to the examiner’s suggestion, there is no experimentation involved at all in selecting the precise algorithm the appellant identified in the specification.

The examiner reads the specification as requiring that “both the positron lifetime algorithm and Doppler-broadening algorithm, each requires two detectors.” Office Action (1/16/07), p. 12; Final Office Action (3/1/06), p. 8. He then states the specification also discloses that one detector may be used. Without reference to any limitations in the claims, the examiner argues that appellant’s “disclosure is thus clearly insufficient and non-enabling

as to which specific known positron lifetime algorithm can be used with only a single detector and, which specific Doppler-broadening algorithm can be used with only a single detector.” Office Action (1/16/07), p. 12; Final Office Action (3/1/06), p. 8. The appellant now understands that “the examiner is not arguing the number of detectors that are enabled by the specification. The issue is the lack of enablement for the specific algorithm to apply when using only a single detector, and the undue experimentation resulting therefrom.” Office Action (1/16/07), p. 19.

Independent claims 1 and 12 claim a “method for evaluating a material specimen, comprising . . . a detector . . . .” However, neither one of these claims contains as a limitation use of a specific algorithm. Thus, once again, the examiner has premised his non-enablement rejections on limitations that are not part of the claims. That is improper and the rejections must be reversed for that reason alone. *CFMT, Inc. v. Yieldup Int’l Corp.*, 349 F.3d 1333, 1338 (Fed. Cir. 2003). (since claims contained no standard for cleanliness, claims for cleaning methods were enabled for any level of contaminant removal); *Ex parte Lemelson*, 2002 WL 32334419, \*4. *See supra* at pp. 16-21.

In addition, the examiner was required to analyze each of the eight required factors set forth in *In re Wands*, 858 F.2d 731, 737 (Fed. Cir. 1988), with reference to the claims, which he did not. Nor did the examiner supply facts to support each and every factor required by *In re Wands*. Even assuming that the examiner had recited any facts (which is denied), merely reciting facts without reference to each of the eight factors is insufficient to sustain a rejection for non-enablement. *Ex parte Kopetzki*, 2001 WL 11197754 (Bd. Pat. App. & Interf. 2001). *See the discussion supra* at pp. 21-23.

Claims 2-10 and 22 ultimately depend from claim 1 and are also allowable at least for the reasons that claim 1 is allowable. Claims 13-19, 21 and 23 ultimately depend from claim 12 and are also allowable at least for the reasons that claim 12 is allowable.

Having failed (1) to base his rejections on limitations in the claims and (2) to analyze the *In re Wands* factors, the examiner failed to meet his prima facie burden to demonstrate that the claims are not enabled. In any event, as the Board previously determined with reference to a related case, descriptions of the various algorithms used in detecting positron annihilation events are sufficiently enabling.

**II. Whether claims 12-19 and 21-23 are unpatentable under 35 U.S.C. §112, first paragraph, as not enabled for failing to include critical elements.**

**A. Legal Standard For Rejection Claims  
Under 35 U.S.C. §112, First Paragraph**

The legal standard set forth above at pages 14-16 is incorporated herein by reference.

In addition, a claim that omits matter disclosed to be essential in the specification may be rejected under §112, first paragraph, as not enabled. *In re Mayhew*, 527 F.2d 1229, 1233 (C.C.P.A. 1976); MPEP 2164.08. However, the specification as a whole must disclose the omitted element or step to be critical to the claimed invention. MPEP 2164.08. As stated in the MPEP, “Features which are merely preferred are not considered to be critical. *In re Goffe*, 542 F.2d 564, 567, 191 USPQ 429, 431 (CCPA 1976).” MPEP 2164.08.

As the MPEP emphasizes, “[A]n enablement rejection based on the grounds that a disclosed critical limitation is missing from a claim should be made only when the language of the specification makes it clear that the limitation is critical for the invention to function as intended. Broad language in the disclosure, including the abstract, omitting an allegedly critical feature, tends to rebut the argument of criticality.” MPEP 2164.08. *See also Ex parte*



*Kato, et al.*, 2002 WL 465390 (Bd. Pat. App. & Interf. 2002) (specification must show the omitted limitation to be critical); *Ex parte Nakano, et al.*, 1998 WL 1735647 (Bd. Pat. App. & Interf. 1998) (same).

B. Response to Examiner's Rejections

The examiner rejected claims 12-19 and 21-23 as not enabled based on his belief that the claims improperly omit the “processing of positron annihilation data by Doppler-broadening algorithm and/or positron lifetime algorithm, which are critical or essential to the practice of the invention.” Office Action (1/16/07), p. 3. However, in making his rejection, the examiner failed to analyze the lack of enablement factors as required by *In re Wands* and he failed to consider the specification as a whole, including disclosures omitting the allegedly critical feature.

a. Claim 21

As an initial matter, the appellant notes that claim 21 contains the limitation “processing the positron annihilation data in accordance with a Doppler-broadening algorithm to produce output data indicative of an accumulation of lattice damage over time.” Since claim 21 already includes the precise limitation that the examiner is arguing needs to be included in the claims as critical, the examiner’s rejection of claim 21 must be reversed.

b. Claims 12-19, 22-23

Turning to the remainder of the claims here rejected, the examiner failed to address the *In re Wands* factors to demonstrate lack of enablement, as required. *See supra* at pp. 14-16. In addition, there is no language in the specification which describes as “critical” or “essential” a step of processing data by Doppler broadening or positron lifetime algorithm. In addition, as described more fully below, the specification (in portions not cited by the

examiner), including the abstract, describe embodiments of the invention that do not include a step for processing data by means of a Doppler broadening or positron lifetime algorithm:

1. The abstract discloses an embodiment of the invention that omits a step of using an algorithm to process data, as follows:

A method for evaluating a material specimen comprises: Mounting a neutron source and a detector adjacent the material specimen; bombarding the material specimen with neutrons from the neutron source to create prompt gamma rays within the material specimen, some of the prompt gamma rays being emitted from the material specimen, some of the prompt gamma rays resulting in the formation of positrons within the material specimen by pair production; collecting positron annihilation data by detecting with the detector at least one emitted annihilation gamma ray resulting from the annihilation of a positron; storing the positron annihilation data on a data storage system for later retrieval and processing; and continuing to collect and store positron annihilation data, the continued collected and stored positron annihilation data being indicative of an accumulation of lattice damage over time.

2. Although the cited example from the abstract demonstrates that the claims at issue are enabled, other embodiments in the specification demonstrate that a step of processing data using an algorithm is not critical either. For example, paragraph 71 discloses an embodiment of the invention for collecting and storing the data, without using an algorithm to process the data, as follows:

[0071] Alternatively, the third embodiment 210 to be used in a long-term arrangement, such as, for example to measure the build-up or accumulation of lattice defects during the service life (or some portion of the service life) of the material specimen 212, the data processing system 224 could be simplified considerably to where the *data processing system 224 serves merely as a data collection device*, collecting the data (e.g., the prompt gamma ray data 220 and/or the positron annihilation data 222) produced by the detector assembly 216, and storing the data in a data story system 225 for later retrieval and processing in accordance with the teachings provided herein. . . . Thereafter, the collected and stored data

*may be* retrieved (i.e., downloaded) and processed in accordance with the teaching herein in order to provide information indicative of the build-up or accumulation of lattice defects during the collection period.

(Emphasis added).

In addition, paragraph 70 of the specification notes that such algorithms *may* (or may not) be used:

[0070] The data processing system 224 is operatively associated with the detector system 216 and receives the prompt gamma ray data 220 and positron annihilation data 222 produced by the detector system 216. As was briefly described above, the data processing system 224 *may* process the prompt gamma ray data 220 and positron annihilation data 222 in accordance with a positron lifetime algorithm (e.g., positron lifetime algorithm 38) . . . [s]o processing the prompt gamma ray data 220 and the positron annihilation data 222 results in positron lifetime data. In addition, the data processing system 224 *may* also process the positron annihilation data 222 in accordance with a Doppler-broadening algorithm (e.g., Doppler-broadening algorithm 40) in the manner already described for the first embodiment 10.

(Emphasis added).

3. The appellant further notes that original claims 12-19 and 22-23 did not contain the allegedly omitted steps. *See* the discussion *infra* at pp. 39-40.

4. As is the case here, where the examiner has failed to provide unequivocal evidence from the specification that the allegedly omitted step was described as essential, the Board has reversed rejections under §112, first paragraph. *See Ex parte Kato*, 2002 WL 465390 at \*5 (“examiner failed to cite any passage of the specification or other statements of record that would establish that any essential element as been omitted from the claim under appeal”). While the examiner here selectively cited from the specification passages to show that algorithms were disclosed in certain embodiments, that disclosure by

itself is not sufficient to establish that critical elements were omitted from the claims, especially where other passages from the specification, including the abstract, describe embodiments without such allegedly omitted elements. *See Ex parte Nakano*, 1998 WL 1735647 at \*4 (“The mere fact that other steps have been disclosed in the preferred embodiment does not render each and every step thereof an essential step.”)

The examiner’s rejections of claims 12-19 and 21-23 under §112, first paragraph, must be reversed.

**III. Whether claims 1-10, 12-19 and 21-23 are patentable under 35 U.S.C. §112, second paragraph, as distinctly claiming the subject matter of the invention.**

**A. Legal Standard For Rejecting Claims  
Under 35 U.S.C. §112, Second Paragraph**

Section 112, second paragraph, requires that the claims particularly point out and distinctly claim what the applicant regards as his invention. 35 U.S.C. § 112. The standard is one of reasonable particularity and completeness. MPEP 2173.02. “The indefiniteness inquiry focuses on whether those skilled in the art would understand the scope of the claim when the claim is read in light of the rest of the specification.” *Union Pacific Res. Co. v. Chesapeake Energy Corp.*, 236 F.3d 684, 691 (Fed. Cir. 2001); *Credle v. Bond*, 25 F.3d 1566, 1576 (Fed. Cir. 1994) (all that is required under § 112, ¶2 is that one of ordinary skill in the art would understand the claim language based on the specification and the drawings). The degree of precision necessary for adequate claims depends on the nature of the subject matter. *Miles Laboratories, Inc., v. Shandon, Inc.*, 27 USPQ2d 1123 (Fed. Cir. 1993). As such, the Patent Office does not permit *per se* rejections of certain language as being indefinite under § 112, ¶ 2. MPEP 2173.02 (“Office policy is not to employ *per se* rules to make technical rejections.”)

The examiner bears the initial burden of presenting evidence or reasoning to explain why persons skilled in the art would not understand the claim language based on the specification. *See In re Oetiker*, 977 F.2d 1443, 1445 (Fed. Cir. 1992). To meet that burden of proof, the MPEP requires that “[d]efiniteness of claim language *must* be analyzed, not in a vacuum, but in light of:

(A) The content of the particular application disclosure;

(B) The teachings of the prior art; and

(C) The claim interpretation that would be given by one possessing the ordinary level of skill in the pertinent art at the time the invention was made.” MPEP 2173.02 (emphasis added). *See In re Johnson*, 558 F.2d 1008, 1015, 194 USPQ 187 (CCPA 1977). In addition, the Federal Circuit has repeatedly held that the enablement requirement and the written description requirement of 112 are not to be conflated. “We interpret 35 U.S.C. §112, ¶1 to require a written description requirement separate and apart from the enablement requirement.” *In re Curtis*, 354 F.3d 1347, 1357 (Fed. Cir. 2004). *See In re Hyatt*, 708 F.2d 712, 715 (Fed. Cir. 1983) (the fact that a claim is not enabled does not render it indefinite). “After evidence or argument is submitted by the applicant in response, patentability is determined on the totality of the record with due consideration to persuasiveness of argument.” *In re Oetiker*, 977 F.2d at 1445.

B. Response to Examiner’s Rejections

The examiner never established a *prima facie* case of indefiniteness. Indeed, the examiner’s rejections here are indefinite, not the appellant’s claim language. In short, by confusing breadth with indefiniteness, the examiner has issued improper rejections under §112, second paragraph. Moreover, even assuming that the examiner did carry his initial

burden (which is denied), the appellant demonstrated that the claim language is definite, which evidence the examiner ignored. The indefiniteness rejections should be reversed.

1. The only specific rejection was traversed.

The only specific claim language that the examiner rejected during prosecution was “at least one” emitted gamma ray, which he rejected as indefinite in his office action of May 10, 2005. Claims previously containing that limitation have all been amended to “a plurality of” emitted gamma rays, thereby traversing the examiner’s indefiniteness rejection. The examiner acknowledged that this rejection had been traversed in his October 17, 2005 office action. Despite this acknowledgement, in the same October 17, 2005 office action, the examiner newly rejected all of the claims as “vague and indefinite” without even specifying which claim language would not be understood by one of ordinary skill in the art. In the final office action, the examiner summarily dismissed the appellant’s argument and evidence regarding definiteness as “not convincing” and merely incorporated by reference his previous action of October 17, 2005 which incorporated his previous action of May 10, 2005 which incorporated his arguments directed to the enablement issue. Similarly, in his non-final office action of January 16, 2007, the examiner rejected the claims as “vague and indefinite” again, with reference to the same October 17, 2005 office action. The examiner’s only response to the appellant’s initial appeal brief was to assert that “appellant’s arguments are: a) simply repetitions of items that have been adequately addressed above.” Office Action (1/16/07), p. 19.

Thus, when reduced to its essence, prosecution in this case has produced only one specific rejection for indefiniteness. The only specific rejection having been traversed, the examiner’s rejection for indefiniteness should be reversed on those grounds alone.

2. The examiner failed to meet his prima facie burden.

Once his initial indefiniteness rejection was traversed, the examiner never specifically and directly identified the claim language that he considered “vague and indefinite.” Rather, as stated above, he just bootstrapped his indefiniteness rejections onto his lack of enablement rejections. The examiner’s indefiniteness rejections are therefore improper.

Not only did he fail to identify any deficient claim language, but also the examiner failed to perform the indefiniteness analyses required by MPEP 2173.02. For example, since the examiner never identified the claim language at issue, he certainly did not interpret the claim language, as required by the MPEP. Further the examiner has not provided any persuasive reasoning why any specific claim language is vague and indefinite.

Even assuming that such language were specified, by relying only on arguments he made to support his non-enablement rejections to support his indefiniteness rejections, the examiner failed to carry his burden. Indeed, the Federal Circuit has specifically stated, “[I]f the ‘enabling’ disclosure of a specification is not commensurate in scope with the subject matter encompassed by a claim, that fact does not render the claim imprecise or indefinite or otherwise not in compliance with the *second* paragraph of §112; rather the claim based on an *insufficient disclosure* (§112, first paragraph) and should be rejected on that ground.” *In re Hyatt*, 708 F.2d at 715.

Having been based entirely on his non-enablement rejections, appellant therefore believes that the examiner’s indefiniteness rejections extend to the scope of the Doppler-broadening algorithm claim limitation of claims 1 and 21, as well as claims 2-10 and 22 which depend from claim 1. (The appellant has already explained the error in the examiner’s non-enablement rejections as improperly based on limitations not found in the claims. *See*

*supra* at pp.16-21). However, the examiner's rejections of all the claims as indefinite cannot be sustained on that basis.

a. Independent claim 12 and dependent claims 13-19 and 23

First, claims 12-19 and 23 do not require the use of any algorithm and the examiner has put forth no evidence or reasoning as to why any specific claim limitation of those claims is imprecise. Having made out no case whatsoever that any language in claims 12-19 and 23 is indefinite, the examiner's rejections in that regard must be reversed.

b. Independent claim 1 and dependent claim 21

Second, with respect to the indefiniteness rejections of claim 1 (and its dependent claims 2-10 and 22) and claim 21 presumably based on the use of a Doppler-broadening algorithm, the words of the Board in the related appeal apply just as well here: "[T]he examiner's criticisms of the claim limitations pertaining to the algorithms . . . focus on the breadth of those limitations. It is well settled, however, that mere breadth does not equate to indefiniteness. In re Miller, 441 F.2d 689, 169 USPQ 597, 600 (CCPA 1971). Accordingly, the examiner's contention that the appealed claims are indefinite is unpersuasive." *Ex parte Akers, supra*, at 9-10.

3. The examiner failed to consider evidence of definiteness.

a. Independent claim 1 and dependent claims 2 and 21

Although the examiner never identified the specific claim language he considered to be indefinite, the applicant surmised that his rejections must have related to the claims language "processing . . . in accordance with a Doppler-broadening algorithm" (claims 1 and 21) and "calculating positron lifetime data" (claim 2). (Neither of these terms is present in independent claim 12 or claims 13-19 and 23). Based on that understanding, the appellant



responded with evidence and argument, explaining why the “various algorithms are not vague [or] indefinite . . . i.e., a the person having ordinary skill in the art would understand the algorithms and possess the knowledge required to evaluate the many algorithms available and select those that would be appropriate for the desired application.” Amendment (1/13/06), p. 13. Such evidence and argument demonstrated that the examiner’s rejections did not meet his prima facie burden.

Specifically, the appellant argued in his Amendment (1/13/06), pp. 12-13 that the claim language was not indefinite because:

(i) Persons having ordinary skill in the art would understand the algorithms and possess the knowledge required to evaluate the many algorithms available and select those that would be appropriate for the desired application.

(ii) With regard to the Doppler-broadening algorithm, the written description clearly states that several different types of Doppler-broadening techniques have been developed and could be used. The written description also states that the Doppler-broadening algorithm may comprise the algorithm disclosed in U.S. Patent No. 6,178,218.

(iii) The written description specifically states that systems for detecting positron lifetimes, as well as the algorithms utilized thereby, are well-known in the art and could be easily provided by persons having ordinary skill in the art after having become familiar with the teachings of the present invention.

Instead of addressing these arguments and presenting evidence why persons of ordinary skill in the art would not understand the meaning the claim terms set forth above, the examiner pronounced the arguments “not convincing.” Final Office Action (3/1/06) at p. 2. Without any further explanation, he merely incorporated his previous rejections by reference

into his final office action. Final Office Action (3/1/06) at p. 12. No clarification was forthcoming in the most recent office action, mailed January 16, 2007, either. In a similar situation where, in the examiner's answer, the examiner responded that the appellants' arguments were "not persuasive," the Board held that the examiner failed to satisfy his burden. *See Ex parte Voordum*, 2002 WL 1801166, \*4 (Bd. Pat. App. & Interf. 2002).

Even assuming that the examiner had specified the claim language set forth above as indefinite (which is denied), one of ordinary skill in the art would well understand the scope of "processing . . . in accordance with a Doppler-broadening algorithm" (claims 1 and 21) and "calculating positron lifetime data" (claim 2) in light of the specification. The examiner has failed to carry his burden to demonstrate otherwise.

Claims 3-10 and 22 depend from claim 1 and are allowable at least for the reasons that claim 1 is allowable.

b. Independent claim 12 and dependent claims 13-19 and 23

As mentioned above, claims 12-19 and 23 contain no limitations directed to any algorithmic claim limitation that the examiner evidently considers indefinite. Therefore, having failed to raise any specter that these claims are indefinite, the examiner's rejections as to these claims should be summarily reversed.

**IV. Whether claims 12-19 and 21-23 are unpatentable under 35 U.S.C. §112, second paragraph, for failing to include critical elements.**

A. Legal Standard for Rejecting Claims Under  
35 U.S.C. §112, Second Paragraph

The legal standard for rejecting claims as indefinite under §112, second paragraph is set forth above at pages 31-32 and is incorporated herein.

In addition, a claim which fails to interrelate essential elements of the invention may

be rejected as indefinite under §112, second paragraph. MPEP 2172.01. However, the Board has cautioned that an “‘incompleteness’ rejection should be extremely rare . . . .” *Ex parte Trummer*, 1997 WL 33106615, \*4 (Bd. Pat. App. & Interf. 1997). The Board explained that this “rejection could be applied in almost every case since it could be said that every part of a disclosed combination is somehow essential to the overall purpose of the invention. This would infringe an applicant’s right to claim what he regards as his invention.” *Id.* The examiner bears the burden of demonstrating by reference to the specification as a whole that the allegedly omitted element is, in fact, critical. *Ex parte Nakano*, 1998 WL 1735647 at \*3. “The mere fact that other steps have been disclosed in the preferred embodiment does not render each and every step thereof an essential step.” *Id.* The fact that an originally filed claim does not include the allegedly omitted elements “indicates the original intent to claim the system without either element.” *Ex parte Trummer*, 1997 WL 33106615, at \*4.

#### B. Response to Examiner’s Rejections

The examiner rejected claims 12-19 and 21-23 “as being incomplete” for failing to include what he believes to be the essential step of “processing the positron annihilation data (e.g., see claim 12) and the prompt gamma ray data (e.g., see claim 13). . . .” Office Action (1/16/07), p. 5. However, the examiner failed to demonstrate that the appellant described the allegedly omitted step as essential.

##### a. Claim 21

As an initial matter, the appellant notes that claim 21 contains the limitation “processing the positron annihilation data in accordance with a Doppler-broadening algorithm to produce output data indicative of an accumulation of lattice damage over time.” Since

claim 21 already includes the precise limitation that the examiner is arguing needs to be included in the claims as a critical step, the examiner's rejection of claim 21 must be reversed.

b. Claims 12-19, 22-23

Turning to the remainder of the claims here rejected, there is no language in the specification which describes as "critical" or "essential" a step of processing data by Doppler broadening or positron lifetime algorithm. In addition, as described more fully below, the specification (in portions not cited by the examiner), including the abstract, describe embodiments of the invention that do not include a step for processing data by means of a Doppler broadening or positron lifetime algorithm. The appellant has set forth above examples from the specification, including the abstract, describing embodiments of the invention that do not include processing positron annihilation data or prompt gamma ray data. *See supra* at pp. 29-30. In addition, the appellant notes that originally filed claims 12-19 and 22-23 did not include a processing step. For example, independent claim 12, as originally filed, claimed:

"A method for evaluating a material specimen, comprising:

mounting a neutron source adjacent the material specimen;

mounting a detector adjacent the material specimen;

bombarding the material specimen with neutrons from the neutron source to create prompt gamma rays within the material specimen, some of the prompt gamma rays being emitted from the material specimen, some of the prompt gamma rays resulting in the formation of positrons within the material specimen by pair production;

collecting positron annihilation data by detecting with the detector at least one emitted annihilation gamma ray resulting from the annihilation of a positron, the detector producing the positron annihilation data;

storing the positron annihilation data on a data storage system for later retrieval and processing; and

continuing to collect and store positron annihilation data, the continued collected and stored positron annihilation data being indicative of an accumulation of lattice damage over time.”

No processing step is included.

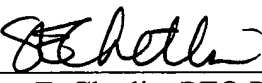
The Board in *Ex parte Trummer*, 1997 WL 33106615, at \*4, found dispositive the fact that an originally filed claim did not include the allegedly omitted element. In that case, the examiner rejected one claim under §112, second paragraph, because that claim did not include controllers, interface module, blockable coupling element, control lines and other structure the examiner regarded as essential. The Board reversed. The Board’s analysis applies equally well in this case: “The Examiner does not point to any evidence that Appellants regard their invention to be something other than what is claimed. The fact that the originally filed independent claim 1 did not include a controller or an interface module indicates an intent to claim the system without either element.” *Id.* Similarly, here, the fact that the abstract, an alternative embodiment disclosed in the specification, and claims 12-19 and 22-23 do not include the allegedly omitted processing step demonstrates that the appellant intended to claim his invention without that step. The examiner therefore has failed to carry his burden of demonstrating that these claims are incomplete. The rejections of these claims under §112, second paragraph, must be reversed.

### **CONCLUSION**

While the examiner argues that the claims here are deficient for being indefinite, non-enabled and lacking critical elements, to the contrary, it is the rejections here that are deficient. Thus, the examiner’s non-enablement rejections were largely based on limitations

not found in the claims. Where the rejections were based on limitations actually found in the claims, the examiner failed to perform the enablement analyses required by *In re Wands, supra*. With respect to indefiniteness, the examiner failed to identify any specific claim language that he considered to be indefinite. Merely piggybacking indefiniteness rejections on non-enablement rejections is improper, as the Federal Circuit has held. None of these deficiencies were cured by the examiner's new omitted element rejections either. The examiner's arguments that certain claim limitations were "critical" and had to be included in the claims were belied by the specification, which disclosed embodiments that did not contain the allegedly omitted step and which did not describe the allegedly omitted limitation as "critical" or "essential." This case has been pending for over three years and has gone in and out of appeal. Being neither indefinite nor non-enabled, these claims are patentable and the appellant is entitled to have them issued.

Respectfully submitted,  
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Date: March 22, 2007

## CLAIMS APPENDIX

1. A method for evaluating a material specimen, comprising:
  - mounting a neutron source adjacent the material specimen;
  - mounting a detector adjacent the material specimen;
  - bombarding the material specimen with neutrons from the neutron source to create prompt gamma rays within the material specimen, some of the prompt gamma rays being emitted from the material specimen, some of the prompt gamma rays resulting in the formation of positrons within the material specimen by pair production;
  - collecting positron annihilation data by detecting with the detector a plurality of emitted annihilation gamma rays resulting from the annihilation of positrons, the detector producing the positron annihilation data;
  - processing collected positron annihilation data in accordance with a Doppler-broadening algorithm; and
  - continuing to collect and process positron annihilation data to measure an accumulation of lattice damage over time.
2. The method of claim 1, further comprising:
  - collecting prompt gamma ray data by detecting with the detector a plurality of emitted prompt gamma rays, the detector producing the prompt gamma ray data;
  - calculating positron lifetime data from the positron annihilation data and the prompt gamma ray data; and

continuing to collect positron annihilation data and prompt gamma ray data and calculate positron lifetime data to measure an accumulation of lattice damage over time.

3. The method of claim 1, wherein said mounting a neutron source adjacent the material specimen comprises mounting the neutron source to the material specimen.

4. The method of claim 3, wherein said mounting a detector adjacent the material specimen comprises mounting the detector to the material specimen.

5. The method of claim 4, further comprising positioning a shield adjacent the neutron source to absorb stray neutrons.

6. The method of claim 5, further comprising positioning a moderator between the neutron source and the material specimen.

7. The method of claim 6, further comprising positioning a reflector adjacent the neutron source to reflect neutrons toward the material specimen.

8. The method of claim 1, wherein mounting a neutron source adjacent the material specimen comprises mounting an isotopic neutron source adjacent the material specimen.



9. The method of claim 8, wherein mounting an isotopic neutron source adjacent the material specimen comprises mounting a neutron source of  $^{252}\text{Cf}$ .

10. The method of claim 1, wherein continuing to collect and process positron annihilation data to measure an accumulation of lattice damage over time is performed while the material specimen is in service.

11. (Canceled).

12. A method for evaluating a material specimen, comprising:

mounting a neutron source adjacent the material specimen;

mounting a detector adjacent the material specimen;

bombarding the material specimen with neutrons from the neutron source to create prompt gamma rays within the material specimen, some of the prompt gamma rays being emitted from the material specimen, some of the prompt gamma rays resulting in the formation of positrons within the material specimen by pair production;

collecting positron annihilation data by detecting with the detector a plurality of emitted annihilation gamma rays resulting from the annihilation of positrons, the detector producing the positron annihilation data;

storing the positron annihilation data on a data storage system for later retrieval and processing; and

continuing to collect and store positron annihilation data, the continued collected and stored positron annihilation data being indicative of an accumulation of lattice damage over time.

13. The method of claim 12, further comprising:  
collecting prompt gamma ray data by detecting with the detector a plurality of emitted prompt gamma rays, the detector producing the prompt gamma ray data;  
storing prompt gamma ray data on the data storage system for later retrieval and processing; and  
continuing to collect and store prompt gamma ray data, the continued collected and stored prompt gamma ray data being indicative of an accumulation of lattice damage over time.
14. The method of claim 12, wherein said mounting a neutron source adjacent the material specimen comprises mounting the neutron source to the material specimen.
15. The method of claim 14, wherein said mounting a detector adjacent the material specimen comprises mounting the detector to the material specimen.
16. The method of claim 15, further comprising positioning a shield adjacent the neutron source to absorb stray neutrons.
17. The method of claim 16, further comprising positioning a moderator between the neutron source and the material specimen.
18. The method of claim 12, wherein mounting a neutron source adjacent the material specimen comprises mounting an isotopic neutron source adjacent the material specimen.

19. The method of claim 12, wherein continuing to collect and store positron annihilation data is performed while the material specimen is in service.

20. (Canceled).

21. The method of claim 12, further comprising:  
retrieving stored positron annihilation data; and  
processing the positron annihilation data in accordance with a Doppler-broadening algorithm to produce output data indicative of an accumulation of lattice damage over time.

22. The method of claim 1, further comprising removing the neutron source before collecting positron annihilation data.

23. The method of claim 12, further comprising removing the neutron source before collecting positron annihilation data.

24. (Canceled).

## EVIDENCE APPENDIX

None.

RELATED PROCEEDINGS APPENDIX

1. *Ex parte Akers*, Appeal No. 2005-0855 (Bd. Pat. App. & Interf. 2005) (slip op.).

The opinion in support of the decision being entered today was not written for publication and is not binding precedent of the Board.



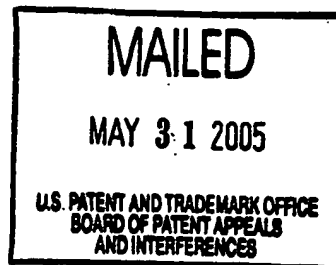
UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS  
AND INTERFERENCES

Ex parte DOUGLAS W. AKERS

Appeal No. 2005-0855  
Application No. 10/269,807

HEARD: May 18, 2005



Before PATE, MCQUADE, and BAHR, Administrative Patent Judges.  
MCQUADE, Administrative Patent Judge.

DECISION ON APPEAL

Douglas W. Akers originally took this appeal from the final rejection (mailed June 16, 2003) of claims 1 and 8 through 39, all of the claims pending in the application. After the appellant's main and reply briefs (filed October 3, 2003 and January 13, 2004) and an examiner's answer (mailed December 1, 2003) had been made of record, the examiner issued an Office action (mailed February 25, 2004) reopening prosecution and entering superseding rejections of all of the claims. Pursuant to 37 CFR § 1.193(b)(2)(ii), the appellant then submitted a request that the appeal be reinstated (filed April, 14, 2004) and

a supplemental brief (filed August 2, 2004). The examiner, implicitly granting the request, entered a second answer (mailed September 14, 2004) and forwarded the application to this Board for review of the new rejections of claims 1 and 8 through 39.

THE INVENTION

The invention relates to "methods . . . for performing non-destructive testing of materials using positron annihilation" (specification, page 1). Representative claim 1 reads as follows:

1. A method, comprising:  
determining whether a specimen to be tested includes at least one positron emitter therein that will be activated in response to photon bombardment;  
selecting a positron emitter to be activated;  
determining a threshold photon energy required to activate the selected positron emitter;  
determining a half-life of the selected positron emitter;  
and  
when the half-life of the selected positron emitter is less than a selected half-life, then performing a rapid activation/analysis process, said rapid activation/analysis process comprising:  
activating for an activation time the selected positron emitter by bombarding the specimen with photons having energies at least as great as the threshold photon energy;  
detecting for a detection time gamma rays produced by annihilation of positrons with electrons in the specimen; and  
repeating said steps of activating for an activation time and detecting for a detection time until detecting a sufficient number of gamma rays to determine at least one material characteristic of said specimen;

when the half-life of the selected positron emitter is greater than or equal to the selected half-life, then performing a normal activation/analysis process, said normal activation/analysis process comprising:

activating the selected positron emitter by bombarding the specimen with photons having energies at least as great as the threshold photon energy; and

detecting gamma rays produced by annihilation of positrons with electrons in the specimen.

#### THE REJECTIONS

Claims 1 and 8 through 39 stand rejected under 35 U.S.C. § 112, first paragraph, as being based on a specification which fails to comply with the enablement requirement.

Claims 1 and 8 through 39 also stand rejected under 35 U.S.C. § 112, second paragraph, as failing to particularly point out and distinctly claim the subject matter the appellant regards as the invention.

Attention is directed to the main, reply and supplemental briefs and the second answer for the respective positions of the appellant and the examiner regarding the merits of these rejections.<sup>1</sup>

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<sup>1</sup> In the Office action which reopened prosecution, claims 1 and 8 through 10 additionally stood rejected under 35 U.S.C. § 112, first paragraph, as being based on a specification which fails to comply with the written description requirement. Upon reconsideration, the examiner has withdrawn this rejection (see page 2 in the second answer). Presumably, the examiner also has withdrawn any reasoning relating to the two remaining rejections which was set forth in the Office action but not restated in the

(continued...)



## DISCUSSION

### I. Preliminary matter

In the supplemental brief (see pages 3, 4 and 17) and second answer (see pages 11 and 12), the appellant and the examiner debate the propriety of the Office action which reopened prosecution subsequent to appeal. As this matter is not directly connected with the merits of any issue involving a rejection of claims, it is reviewable by petition to the Director rather than by appeal to this Board (see In re Hengehold, 440 F.2d 1395, 1403-04, 169 USPQ 473, 479 (CCPA 1971), and hence will not be further addressed in this decision.

### II. The 35 U.S.C. § 112, first paragraph, rejection

The dispositive issue with respect to the enablement requirement of 35 U.S.C. § 112, first paragraph, is whether the appellant's disclosure, considering the level of ordinary skill in the art as of the date of the application, would have enabled a person of such skill to make and use the claimed invention without undue experimentation. In re Strahilevitz, 668 F.2d 1229, 1232, 212 USPQ 561, 563-64 (CCPA 1982). In calling into

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<sup>1</sup>(...continued)  
second answer (see Ex parte Emm, 118 USPQ 180, 181 (Bd. App. 1957)).

question the enablement of the disclosure, the examiner has the initial burden of advancing acceptable reasoning inconsistent with enablement. Id.

The examiner considers the appellant's disclosure to be non-enabling in four respects:

. . . There is neither an adequate description nor enabling disclosure as to what is encompassed by the term, "activating a positron emitter". A "positron emitter" is inherently already activated, i.e., it is radioactive.

At best, the use of the term "activating a positron emitter" is superfluous.

At worst, the term would imply that a positron emitter is either being transformed to another positron emitter or its energy level is further raised by the energy of the activating photon. There is no support in the specification for either one of these two alternatives [second answer, page 3];

There is neither an adequate description nor enabling disclosure as to how and in what manner potential interferences in the data are accounted for in the analysis. For example, Claim 1 recites the step of bombarding the specimen with photons at least as great as the threshold photon energy required to activate the selected positron emitter and detecting gamma rays produced by annihilation of positrons with electrons in the specimen.

. . . [T]here is no support as to how one would differentiate between the signals from the selected positron emitter and from the non-selected ones [second answer, pages 6 and 7];

There is neither an adequate description nor enabling disclosure as to how and in what manner one should select an algorithm from a plurality of available algorithms, modify/manipulate the selected algorithm and evaluate the constants in the selected

algorithm to fit Appellant's situation. Appellant himself admits in the specification that there is not only one known algorithm but rather a plurality of known positron lifetime algorithms (e.g. see paragraph 0058), Doppler broadening algorithms (paragraph 0057) and three-dimensional imaging algorithms (e.g., see paragraph 0060).

. . . There is no support as to how and in what manner one selects the specific algorithm to use for the two activation/analysis processes, to evaluate requisite constants and to modify the selected algorithm to Appellant's situation [second answer, page 7];

and

There is neither an adequate description nor enabling disclosure as to how and in what manner one can determine when the half-life of the selected positron emitter is less than a selected half-life (sic), where the composition of the specimen is unknown and it is this composition that is desired to be determined. The same lack of support exists for the determination of when the half-life of the selected positron emitter is more than a selected half-life (sic) [second answer, page 8].

A fair assessment of the appellant's disclosure shows that the references therein to "activating" a positron emitter through photon bombardment of the specimen would be understood by the artisan as synonymous with "producing" or "forming" the positron emitter from a stable precursor. For example, the specification states that "photons 16 from the photon source 12 produce such neutron-deficient isotopes . . . referred to herein in the alternative as 'positron emitters'" (page 7, emphasis added), and

that "one way for generating positrons is through the formation within the specimen 18 of neutron-deficient isotopes, i.e., positron emitters" (page 20, emphasis added). In the same vein, the specification describes Tables I and II (on pages 21 and 22) as identifying "those isotopes that may be converted into positron emitters by photon bombardment" (page 20, emphasis added). Thus, even if the appellant's description of "activating" a positron emitter is somewhat inaccurate as urged by the examiner, it is so only in the most hyper-technical sense. Read in context, the appellant's terminology would not pose any enablement problem from the perspective of a person having ordinary skill in the art.

The examiner's concerns as to possible interference between respective signals produced by the selected positron emitter and non-selected positron emitters and as to the half-life of the selected positron emitter also are unfounded, primarily because they are not commensurate with the scope of the invention disclosed and claimed by the appellant. In this regard, the claims do not exclude the activation of multiple positron emitters, and indeed the appellant's specification (see pages 8, 13 and 14) actually allows for such activation. The claims also

do not require the composition of the specimen to be unknown, and are not directed to a method for determining the composition of the specimen.

Finally, while the appellant's disclosure does not convey much detail as to the various algorithms described therein, these algorithms admittedly are generally known in the art. The examiner has failed to advance any cogent reasoning as to why the disclosure would not have enabled a person having ordinary skill in the art to employ these algorithms, without undue experimentation, to the extent required by the appealed claims.

Hence, the examiner's position that the appellant's disclosure, considering the level of ordinary skill in the art as of the date of the application, would not have enabled a person of such skill to make and use the claimed invention without undue experimentation is not well taken. Therefore, we shall not sustain the standing 35 U.S.C. § 112, first paragraph, rejection of 1 and 8 through 39.

III. The 35 U.S.C. § 112, second paragraph, rejection

The second paragraph of 35 U.S.C. § 112 requires claims to set out and circumscribe a particular area with a reasonable degree of precision and particularity. In re Johnson, 558 F.2d

1008, 1015, 194 USPQ 187, 193 (CCPA 1977). In determining whether this standard is met, the definiteness of the language employed in the claims must be analyzed, not in a vacuum, but always in light of the teachings of the prior art and of the particular application disclosure as it would be interpreted by one possessing the ordinary level of skill in the pertinent art. Id.

The examiner views the appealed claims to be indefinite for reasons essentially similar to those listed above in connection with the enablement rejection. In this regard, the examiner points to the claim limitations relating to (1) the activation of a positron emitter, (2) the positron lifetime, Doppler broadening, three-dimensional imaging and activation/analysis process algorithms and (3) the determination of the half-life of the selected positron emitter (see pages 9 through 11 in the second answer). For the reasons explained previously, the limitations pertaining to the activation of a positron emitter would have been readily understood by the artisan when read in light of the underlying specification. Furthermore, the examiner's criticisms of the claim limitations pertaining to the algorithms and the half-life of the selected positron emitter

focus on the breadth of these limitations. It is well settled, however, that mere breadth does not equate to indefiniteness.

In re Miller, 441 F.2d 689, 169 USPQ 597, 600 (CCPA 1971).

Accordingly, the examiner's contention that the appealed claims are indefinite is unpersuasive.

Thus, we shall not sustain the standing 35 U.S.C. § 112, second paragraph, rejection of claims 1 and 8 through 39.

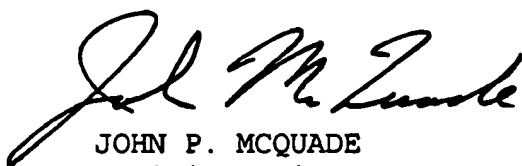
SUMMARY

The decision of the examiner to reject claims 1 and 8 through 39 is reversed.

REVERSED



WILLIAM F. PATE, III  
Administrative Patent Judge



JOHN P. MCQUADE  
Administrative Patent Judge



JENNIFER D. BAHR  
Administrative Patent Judge

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Appeal No. 2005-0855  
Application No. 10/269,807

Page 12

ALAN D. KIRSCH  
P. O. BOX 1625  
IDAHO FALLS, ID 83415-3899



## REFERENCES APPENDIX

Copies of the following references are attached hereto for the Board's convenience:

1. U.S. Patent No. 4,064,438 issued to Alex, et al.
2. U.S. Patent No. 6,178,218, issued to Akers, et al.
3. U.S. Patent No. 7,058,153, issued to Akers.
4. Y. Zhu & R. Gregory, "Analysis of Positron Annihilation Lifetime Data Presented as a Sum of Convolved Exponentials with the Program "SPLMOD," *Nuclear Instruments & Methods in Physics Research A284*, 443-451 (1989).
5. J. Schaffer, "Deconvoluted Doppler Broadened Positron Annihilation Spectroscopy: Characterization of Defects in Aluminum," Ph.D. Thesis, Duke University, University Microfilms International (1985).
6. P. Derlet, T. Choy, "A positron annihilation lifetime spectroscopy study of porous silicon using a continuous lifetime fitting algorithm," *Journal of Material Science Letters 15*, 1949-1952 (1996).
7. P. Bandžuch, M. Morháč, J. Krištiak, "Study of Van Cittert and Gold iterative methods of deconvolution and their application in the deconvolution of experimental spectra of positron annihilation," *Nuclear Instruments & Methods in Physics Research A384*, 506-515 (1997).

## ANALYSIS OF POSITRON ANNIHILATION LIFETIME DATA PRESENTED AS A SUM OF CONVOLUTED EXPONENTIALS WITH THE PROGRAM "SPLMOD"

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Received 12 December 1988 and in revised form 12 June 1989

The reliability of a least squares analysis of positron annihilation lifetime data represented as a sum of convoluted exponentials is investigated with computer-stimulated test data. The method of analysis, which is available as a computer program called "SPLMOD" (R.H. Vogel, Technical Report EMBL-DA08, European Molecular Biology Laboratory, Heidelberg, FRG, (1986)), avoids direct determination of the instrument resolution function by employing the decay curve of a reference material with a well-known single lifetime. The performance of the algorithm for extracting positron annihilation lifetime information was evaluated by using several measures of the information content of the decay curve described recently by Schrader and Usmar [in: *Positron Annihilation Studies of Fluids*, ed. S. Sharma (World Scientific, Singapore, 1988)]. The sensitivity of the algorithm to systematic errors including errors in the resolution function, shifts in the zero time channel and contamination of the reference decay curves by additional lifetime components was evaluated. The method provides excellent estimates of lifetime parameters when the reference decay curve accurately reflects the resolution function of the sample data. However, the algorithm is extremely sensitive to errors in the instrument resolution function.

### 1. Introduction

The analysis of positron annihilation lifetime data is an extremely difficult undertaking. The decay curve is usually represented by:

$$C(t) = \sum_{i=1}^n \alpha_i \lambda_i \exp(-\lambda_i t), \quad (1)$$

where  $\alpha_i$  is the number of positrons annihilating with natural lifetime,  $\lambda_i^{-1}$ . Analysis of sums of exponentials is notoriously difficult, particularly when the number of components,  $n$ , is unknown. In general, even for data with arbitrarily small noise levels, there exist a large number of solutions,  $\{\alpha_i, \lambda_i, n\}$ , that fit the data to within the noise level. These difficulties are compounded by the fact that in a positron annihilation lifetime experiment one cannot observe the function  $C(t)$  directly, only its convolution with the instrument resolution function,  $R(t)$ :

$$y(t) = R(t) * C(t). \quad (2)$$

The difficulties associated with the analysis of eq. (2) are therefore two-fold: the choice of an appropriate representation of  $C(t)$  and the determination of the function,  $R(t)$ .

The problems associated with analysis of data with eq. (1) are very well known. In general, we have no prior knowledge of the number of components,  $n$ , in eq. (1). If  $n$  is too large the solutions become very unstable to noise in the data. Because of this difficulty, it is common to assume only two or three components in eq. (1). This has the effect of stabilizing the solution to noise and in some simple systems may provide a physically meaningful representation of the decay. However, in more complex systems setting  $n$  too small will result in an inadequate representation of the system, resulting in a loss of valuable information and a set of  $\alpha_i$  and  $\lambda_i$  pairs that may have little or no physical meaning. Because of the non-linearity of  $C(t)$  in the  $\lambda_i$ , it is not possible to extend linear hypothesis tests to define an upper value for  $n$ . However, Provencher [1] has described a hypothesis test which incorporates a correction for non-linearity that has been found to be very effective. It is employed in the programs DISCRETE, which solves sums of exponentials and SPLMOD which solves more general sums of one parameter functions, including exponentials and convoluted exponentials. The use of eq. (1) assumes that a discrete sum of exponentials is a physically meaningful representation of the data. For many simple, homogeneous systems this is

indeed the case, however, for complex, heterogeneous systems and systems in which distinct structural domains persist for times that are long relative to positron lifetimes, the lifetimes spectrum is expected to be more complex and may consist of a continuous distribution of lifetimes over some interval. For such systems an alternative approach is to replace the sum in eq. (1) by an integral [2-4]:

$$C(t) \sim \int_0^{\infty} \lambda \alpha(\lambda) \exp(-\lambda t) d\lambda = \mathcal{L}\{\lambda \alpha(\lambda)\}, \quad (3)$$

$C(t)$  is simply the Laplace transform of  $\lambda \alpha(\lambda)$ , where  $\alpha(\lambda)$  is the annihilation rate probability density function (pdf). The fraction of positrons annihilating with rates between  $\lambda$  and  $\lambda + d\lambda$  is then given by  $\alpha(\lambda) d\lambda$  (assuming  $\alpha(\lambda)$  is appropriately normalized). In principle,  $\alpha(\lambda)$  can be recovered by inverse Laplace transformation of  $C(t)$ . The problems of solution uniqueness and stability to noise associated with analysis of eq. (1) also plague the inversion of eq. (3). However, recent advances in the solution of Fredholm integral equations provide a number of reliable inversion methods [4-9].

Schrader [2,3] has pioneered the application of Laplace transform methods in the analysis of positron annihilation lifetime data and the application of information theory approaches to the determination of the information content of the experimental data. A particularly important result of this work is the expression of the information content (Shannon Number) of the data in terms of spectrometer parameters (i.e. the number of channels, the channel width, FWHM and the total number of annihilation events observed). Not only does this knowledge define the complexity of the function,  $C(t)$ , that is required to properly represent the data, but it also serves as a guide in the design of experiments to maximize the information content of the data.

The accurate determination of the instrument resolution function,  $R(t)$ , is a major problem in analysis of positron lifetime data. Probably the most widely used method is to determine  $R(t)$  from the prompt spectrum of a  $^{60}\text{Co}$  source. However, the resolution function is very dependent on the distribution of energy deposited in the scintillators. As a result, the function determined with the 1.17 MeV and 1.33 MeV coincident  $\gamma$ -rays from  $^{60}\text{Co}$  will not correctly represent the resolution function when data are collected with a  $^{22}\text{Na}$  source. One approach, that attempts to overcome these difficulties, is to determine the resolution function from the lifetime data themselves. An analytical function, such as a sum of Gaussians or a modified Gaussian, is assumed for  $R(t)$  and its parameters are treated as variables to be determined along with the lifetime parameters of interest [10,11]. However, there are several fundamental objections to this approach. The addition of further parameters to be determined by the analysis can severely

compound the problem of solution uniqueness. In addition, the approach requires the choice of an analytical function to represent the resolution properties of the instrument. An incorrect choice will, of course, transmit errors to the lifetime estimates.

The resolution function may also be determined in a separate experiment from the decay curve of a standard sample with a known single lifetime, such as a well-annealed metal [12]. In principle, this method can yield very good estimates of the instrument resolution function. However, in practice, annihilation in the source and positron trapping at the surface can give rise to additional components. An alternative approach is to employ a  $^{207}\text{Bi}$  source.  $^{207}\text{Bi}$  emits two  $\gamma$ -rays separated by a time interval of 186 ps, the energies of which are almost identical to those employed in a positron lifetime experiment with a  $^{22}\text{Na}$  source. Although the method does not introduce unwanted source components, it is not yet clear if the energies of the  $\gamma$ -rays emitted by  $^{207}\text{Bi}$  are sufficiently similar to those occurring with  $^{22}\text{Na}$  that the types of error associated with the use of  $^{60}\text{Co}$  are entirely avoided.

At present, the use of standard samples with  $^{22}\text{Na}$  sources appears to offer the best hope for reliable determination of the instrument resolution function. We have been interested in the use of a deconvolution method due to Gauduchon and Wahl [13] and Wijnaendts van Resandt et al. [14] for this purpose. A preliminary report of its use in the analysis of positron annihilation lifetime data with numerical Laplace inversion methods has appeared [4] but no tests of its performance have been presented. Our purpose in this paper is to explore the reliability of this deconvolution method in the analysis of positron annihilation lifetime data. The efficiency of the algorithm for extracting lifetime information is evaluated by using several measures of the information content of the decay curve described by Schrader and Usmar [3]. In this work,  $C(t)$  is represented by a sum of exponential terms as in eq. (1) and the analysis is performed with the FORTRAN program, SPLMOD, developed by Vogel [15]. Although eq. (1) is more restrictive than eq. (3), it can be a valid representation of annihilation lifetimes in simple, homogeneous systems and is considerably simpler for our purpose because the measure of information content of the data, the Shannon Number, is directly related to the number of components in the lifetime spectrum.

## 2. Deconvolution of the instrument resolution function

Deconvolution of the instrument resolution function follows the method of Gauduchon and Wahl [13] and Wijnaendts van Resandt et al. [14], which avoids the direct determination of  $R(t)$  by measuring,  $y_r(t)$ , the

decay curve of a reference material with a well-known single lifetime,  $\lambda_r$ :

$$y_r(t) = R(t) * C_r(t), \quad (4)$$

where

$$C_r(t) = \alpha_r \lambda_r \exp(-\lambda_r t). \quad (5)$$

Convoluting eq. (2) with  $C_r(t)$  we obtain:

$$C_r(t) * y(t) = y_r(t) * C(t). \quad (6)$$

Here we assume that  $y_r(t)$  and  $y(t)$  have the same resolution function,  $R(t)$ . This would be true if the reference and sample decay curves could be measured simultaneously, but this is not possible in a positron annihilation lifetime experiment. Any instrument drift occurring between measurement of the sample and reference decay curves will therefore introduce error into the lifetime estimates. This, of course, is true of all methods that involve a separate measurement of the resolution function of the instrument and can only be avoided by improving the stability of the instrument.

Eq. (6) can be rewritten as:

$$\alpha_r \lambda_r \{\exp(-\lambda_r t)\} * y(t) = y_r(t) * C(t), \quad (7)$$

which on taking Laplace transforms gives:

$$\frac{\alpha_r \lambda_r}{\lambda_r + p} \tilde{y}(p) = \tilde{y}_r(p) \tilde{C}(p), \quad (8)$$

where the tilde denotes Laplace transformation and  $p$  is the Laplace variable. Eq. (8) is solved for  $\tilde{y}(p)$ :

$$\tilde{y}(p) = \alpha_r^{-1} \lambda_r^{-1} \{ \lambda_r \tilde{y}_r(p) \tilde{C}(p) + p \tilde{y}_r(p) \tilde{C}(p) \}, \quad (9)$$

which on inverting Laplace transforms gives:

$$y(t) = \alpha_r^{-1} \lambda_r^{-1} \{ \lambda_r y_r(t) * C(t) + C(t) * y_r(t) \}. \quad (10)$$

If  $C(t)$  is represented by eq. (1), eq. (10) becomes:

$$y(t) = \sum_{i=1}^n \frac{\alpha_i \lambda_i}{\alpha_r \lambda_r} \{ y_r(t) + (\lambda_r - \lambda_i) y_r(t) * \exp(-\lambda_i t) \}. \quad (11)$$

If  $\lambda_r$  is known and  $\alpha_r$  is absorbed into the  $\alpha_i$ , eq. (11) must be solved for the  $2n$  parameters  $\alpha_i$  and  $\lambda_i$ . If  $\lambda_r$  is not known, there are  $2n + 1$  parameters.

### 3. Analysis of sums of convoluted exponentials with "SPLMOD"

SPLMOD analyzes data that can be represented as a sum of one parameter functions:

$$y_k = \sum_{i=1}^n \beta_i f(\lambda_i, t_k) + \sum_{j=1}^{n_L} \gamma_j g_j(t_k). \quad (12)$$

The current version of SPLMOD (version 3) distributed by Vogel [15] can perform the following types of analysis of relevance to Positron Annihilation Lifetime (PAL) spectroscopy:

(a) Solution of eq. (12) when the resolution function is known; i.e. with  $f(\lambda_i, t_k) = R(t_k) * \exp(-\lambda_i t_k)$  and  $g_1(t_k) = 1$  (to account for the background).

(b) Solution of eq. (12) using a reference material with a known single annihilation lifetime,  $\lambda_r$ , i.e. with  $f(\lambda_i, t_k) = (\lambda_r - \lambda_i) y_r(t_k) * \exp(-\lambda_i t_k + y_r(t_k))$  and  $g_1(t_k) = 1$ .

(c) Solution of eq. (12) using a reference material with a single annihilation lifetime which need not be known; i.e. with  $f(\lambda_i, t_k) = y_r(t_k) * \exp(-\lambda_i t_k)$ .

$$g_1(t_k) = 1 \text{ and } g_2(t_k) = y_r(t_k).$$

We have employed option (b) in our simulations because it provides more stable solutions than option (c).

SPLMOD performs a weighted least squares analysis in which  $f(\lambda_i, t_k)$  is approximated by a sum of normalized cubic B-splines. The program employs Gauss-Newton minimization, with an extensive search of parameter space to determine the initial parameter values. The convolution,  $y_r(t_k) * \exp(-\lambda_i t_k)$ , is calculated by using a recursive formula of Grinvald and Steinberg [16]:

$$f(\lambda_i, t_{k+1}) = \{ f(\lambda_i, t_k) + 0.5 \Delta t y_r(t_k) \} \times \exp(-\lambda_i \Delta t) + 0.5 \Delta t y_r(t_{k+1}), \quad (13)$$

where  $\Delta t$  is the channel width of the multi-channel analyzer in which the data are collected. SPLMOD performs several analyses in which  $n$  in eq. (1) takes the values: 1, 2, 3, ...,  $n_{\max}$ , where  $n_{\max}$  is set by the user. SPLMOD supplies a number of statistical measures, including Fisher probabilities and random runs probabilities, to aid in the choice of an appropriate solution. Measures of the information content of the data may also be employed to define an appropriate solution [3].

Note that it is not necessary to independently define the zero time point of the spectrometer. This information is already contained in the reference decay curve. However, there is an option in SPLMOD to correct for small changes in the zero time point due to instrument drift occurring between measurement of sample and reference decay curves.

### 4. Simulation of decay curves

Positron annihilation decay curves have been simulated by convoluting eq. (1) with a Gaussian resolution function with FWHM equal to  $2\sigma\sqrt{\ln 2}$ :

$$R(t) = \frac{1}{\sigma\sqrt{\pi}} \exp(-(t/\sigma)^2). \quad (14)$$

This convolution is given by:

$$y(t) = 0.5 \sum_{i=1}^n a_i \lambda_i \exp \left\{ -\lambda_i t + (\lambda_i \sigma / 2)^2 \right\} \times \operatorname{erfc}(\lambda_i \sigma / 2 - t / \sigma). \quad (15)$$

where  $\operatorname{erfc}$  is the compliment of the error function:

$$\operatorname{erfc}(x) = 1 - \operatorname{erf}(x) = 1 - \frac{2}{\sqrt{\pi}} \int_0^x \exp(-t^2) dt. \quad (16)$$

Conversion of the continuous function,  $y(t)$ , to a table of channel counts,  $y_k$ , was performed by numerical integration. A background and appropriate Poisson noise were added to each channel. The reference data were simulated in the same way except that no background was added. The reference and sample spectra were calculated over the same range of channels in each case.

### 5. Information content of the simulated data

Schrader and Usmar [3] have given a detailed discussion of the determination of the information content (Shannon Number) of decay curves and have provided an approximate expression for the Shannon Number in terms of instrument setting of the spectrometer. The Shannon number, in the context of PAL spectroscopy, is a measure of the number of resolvable lifetimes that can be geometrically fitted between the largest and smallest resolvable lifetimes. This measure is an approximation because it depends on ones definition of the relative scatter in the decay curve and on the values that are chosen for the largest and smallest resolvable lifetimes. Schrader and Usmar [3] define the Shannon Number,  $F$ , as:

$$F = \pi^{-1} \omega_{\max} \ln(a_1 N_{\text{ch}} \Delta t / a_2 \text{ FWHM}), \quad (17)$$

where  $a_1$  and  $a_2$  are constants, FWHM is the full width at half the peak height of the resolution function,  $N_{\text{ch}}$  is the number of channels to the right of the zero-time channel,  $N_0$ , employed in the analysis (a maximum value for  $N_{\text{ch}}$  is defined by the number of channels for which the count is greater than the square-root of the background count) and  $\omega_{\max}$  is defined by:

$$\omega_{\max} = 2\pi^{-1} \ln(\sqrt{2\pi} / \eta), \quad (18)$$

$\eta$  is a measure of the effective relative noise in the decay curve and is defined as:

$$\eta = \frac{\sum_{k=N_0}^{N_{\text{ch}}} y_k^{1/2}}{\sum_{k=N_0}^{N_{\text{ch}}} y_k}, \quad (19)$$

$\omega_{\max}$  has a special significance in the eigenfunction expansion of Laplace transforms which we need not be concerned with here [3,5]. Eq. (18) is derived by assuming that the shortest resolvable lifetime is equal to

$a_2$  FWHM and the longest resolvable lifetime is equal to  $a_1 N_{\text{ch}} \Delta t$ . A resolution factor,  $p$ , is defined as the ratio of resolvable lifetimes and it follows that  $F$  is the number of resolvable lifetimes that can be geometrically fitted between  $a_2$  FWHM and  $a_1 N_{\text{ch}} \Delta t$ . The constants  $a_1$  and  $a_2$  need to be determined. In practice, the shortest resolvable lifetime is probably less than FWHM and the longest resolvable lifetime is longer than  $N_{\text{ch}} \Delta t$ , so  $a_2 < 1$  and  $a_1 > 1$ . Schrader and Usmar [3] suggest  $a_1 = 3$  and  $a_2 = \frac{1}{3}$  as reasonable values when  $a_2 \text{ FWHM} > 3\Delta t$ , but they suggest replacing  $a_2 \text{ FWHM}$  by  $3\Delta t$  if  $a_2 \text{ FWHM} < 3\Delta t$ .

The resolution factor is given by:

$$p = \exp(\pi / \omega_{\max}) \quad (20)$$

and is therefore dependent on the definition of the effective relative noise of the decay curve (eq. (19)). Estimates of  $a_1$ ,  $a_2$  and the correctness of eq. (19) are discussed below.

### 6. Analysis of simulated data

Table 1 lists the results of an analysis of simulated PAL data together with the information parameters for the decay curve and will serve as a point of reference for our subsequent discussions. The simulated data contain three components. The reference has a single lifetime of 125 ps. A total of 800 channels were analyzed, starting 30 channels (0.75 ns) to the left of the peak. A plot of

Table 1  
Calculated information content of simulated data and results of its analysis with SPLMOD

FWHM	290 ps	$p$	2.6
$\Delta t$	25 ps	$\eta$	0.015
Reference lifetime	125 ps	$\omega_{\max}$	3.25
Total events	$10^6$	maximum $N_{\text{ch}}$	442

$a_1$	$a_2$	$F$	Resolvable lifetimes (ns)	
			min.	max.
1	1	3.8	0.29	11
2	0.5	5.2	0.15	22
3	0.33	6.0	0.1	33

Lifetimes (ns)			
Simulated	0.125	0.400	2.000
Fitted	0.124	0.397	1.997
		Alpha	
Simulated	0.130	0.470	0.400
Fitted	0.125	0.474	0.401
		Background	
Simulated		100	
Fitted		100.5	

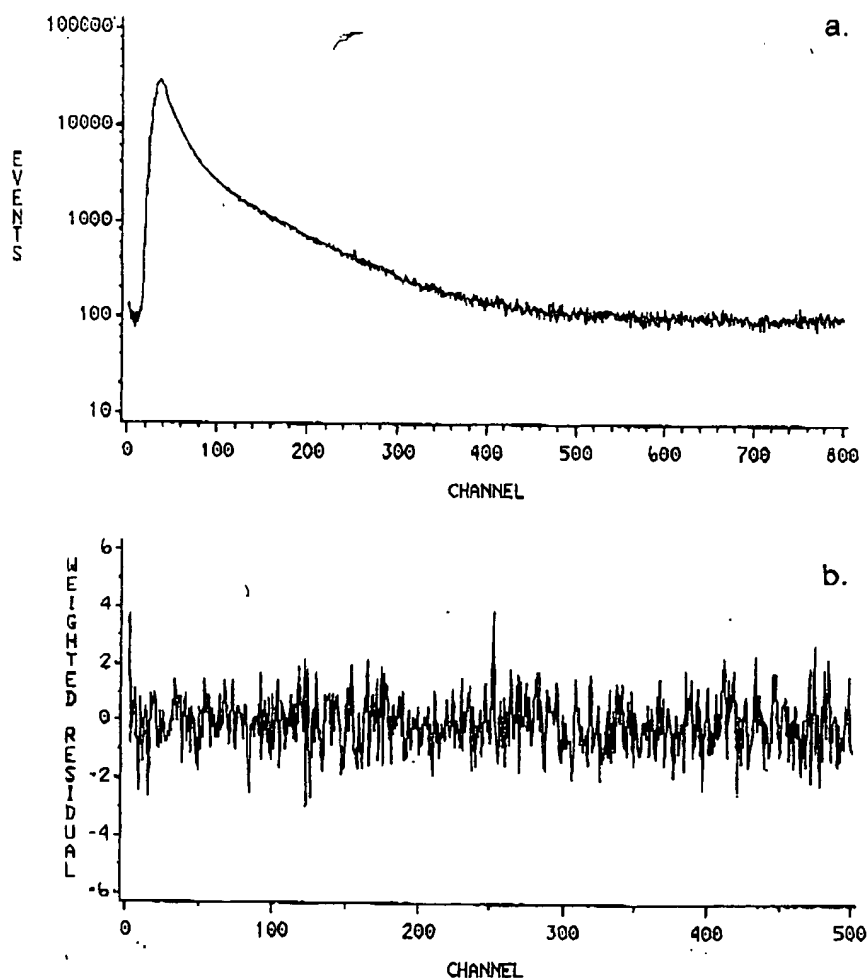


Fig. 1. Example of the fit to the simulated data obtained with SPLMOD. (a) Simulated decay curve calculated with the lifetime parameters given in table 1. (b) Weighted residuals about the zero time channel for the fit given in table 1.

the simulated data and the residuals is shown in fig. 1. The fit to the data is clearly excellent with no evidence of any oscillations of the residuals about the peak. The smallest ratio of simulated lifetimes is 3.2, while the calculated resolution factor is 2.6 SPLMOD has little difficulty in resolving the three components, and provides good estimates of the parameters for the short-lived component (125 ps). The information content of the data, measured by the Shannon number, is clearly sufficient to define all the components even with  $a_1$  and  $a_2$  assumed equal to one.

These results already provide some clues as to the appropriate value for  $a_2$ . It must be  $\leq 0.33$  because the short-lived component is well defined. Table 2 lists the results obtained when FWHM is increased with  $\Delta t = 12.5$  ps. The range of channels employed for analysis was chosen such that the first and last channels were

Table 2

The effect of FWHM on the resolution of lifetime components. Simulation parameters are the same as given in table 1 except that  $\Delta t = 12.5$  ps

		Simulated data	FWHM		
			500 ps	600 ps	700 ps
Lifetime (ns)	1	0.125	0.121	0.116	0.122
	2	0.400	0.400	0.398	0.326
	3	2.000	2.000	1.998	1.633
Alpha	1	0.130	0.129	0.125	0.069
	2	0.470	0.471	0.475	0.526
	3	0.400	0.399	0.400	0.406

sufficient to sample the baseline on both sides of the peak. Good estimates of the parameters are obtained even with FWHM values as large as 500 to 600 ps

( $a_2 = 0.25$  to  $0.20$ ). Beyond this value, SPLMOD is still able to identify three components but the parameter values become very poor. Increasing the total count to  $10^7$  does not improve these estimates. At a FWHM of 290 ps, a value that is typical of modern PAL spectrometers employing plastic scintillators, the shortest lifetime that can be resolved by SPLMOD is approximately 50 ps (i.e.  $a_2 = 0.17$ ). An estimate of  $a_1$  was

determined by varying the number of channels employed in the analysis. In all cases the start channel was sufficiently far to the left of the peak ( $-0.74$  ns) to sample the baseline. With  $N_{ch} = 1940$ , the longest lifetime that can be resolved is approximately 100 ns ( $a_1 = 4$ ).

We have examined the ability of the program to resolve lifetime components and were particularly inter-

Table 3

The resolution of lifetime components with SPLMOD. Variations in the effective relative noise and resolution factor were generated by changing the total number of events. Simulation parameters were; FWHM = 290 ps,  $\Delta t = 12.5$ , reference lifetime = 125 ps and background = 100

	Simulated	Fitted	Fitted	Fitted
Total events		$10^6$	$10^5$	$10^4$
Relative noise		0.024	0.054	0.074
$\omega_{max}$		3.0	2.4	2.4
Resolution factor		2.9	3.6	4.1
$F(a_1 = 4, a_2 = 0.25)$		6.1	4.5	2.7
Maximum $N_{ch}$		959	451	64
Lifetime 1	0.125	0.122	0.113	0.124
(ns) 2	0.400	0.386	0.313	0.609
3	1.280	1.213	0.895	2.786
4	4.096	3.947	3.338	—
Alpha 1	0.250	0.245	0.223	0.341
2	0.250	0.247	0.210	0.345
3	0.250	0.248	0.258	0.314
4	0.250	0.260	0.310	—

Table 4

The effect of systematic errors on lifetime parameters estimated by SPLMOD. Simulation parameters were FWHM = 290 ps,  $\Delta t = 12.5$  ps, background = 100, total events =  $10^7$  and reference lifetime = 125 ps

	Simulated data	Incorrect reference FWHM		Zero channel shift
		285 ps	295 ps	
Lifetime 1	0.125	0.127	0.138	0.134
(ns) 2	0.400	0.409	0.410	0.409
3	2.000	2.004	2.004	2.005
4	—	0.003	100.0	0.002
Alpha 1	0.130	0.663	0.148	0.247
2	0.470	0.171	0.449	0.402
3	0.400	0.150	0.395	0.351
4	—	0.016	0.008	0.001
Background	100	100.4	91.1	99.9
	Simulated data	Contamination of reference by additional component with lifetime = 450 ps		
		5	10	15
Lifetime 1	0.125	0.122	0.120	0.118
(ns) 2	0.400	0.382	0.365	0.347
3	2.000	2.001	2.003	2.005
Alpha 1	0.130	0.131	0.135	0.137
2	0.470	0.476	0.474	0.477
3	0.400	0.393	0.391	0.386
Background	100	100.3	100.2	100.1



tested in determining if the value of the resolution factor calculated according to eq. (20) correctly predicted the point at which SPLMOD failed to resolve lifetime components. An example of the results for such a test is shown in table 3. The simulated data consisted of four components with successive lifetimes increasing by a factor of 3.2. When the total number of events is  $10^6$ , the calculated resolution factor is 2.9 and the Shannon number,  $F$ , is 6.1 SPLMOD is able to resolve the four components and provides reasonably good estimates of the lifetime parameters. When the total number of events is reduced to  $10^5$ , the effective relative noise is doubled, the resolution factor increases to 3.6 and the Shannon number decreases to 4.5. SPLMOD is still able to resolve four components but the lifetime parameter values become poor. When the total number of events is reduced to  $10^4$ , the relative noise increases to 0.074, the

resolution factor increases to 4.1 and the Shannon Number decreases to 2.7. SPLMOD is unable to resolve the four components. Eq. (19) thus appears to provide a good estimate of the effective relative noise in the data as measured by the predictive power of the calculated resolution factor.

## 7. Systematic errors

We have examined the sensitivity of the SPLMOD algorithm to various systematic errors in the data. The most important sources of systematic error arise from: (i) variations in the resolution function for the reference and sample data, (ii) differences in the zero time point for the reference and sample, and (iii) contamination of the reference with additional lifetime components.

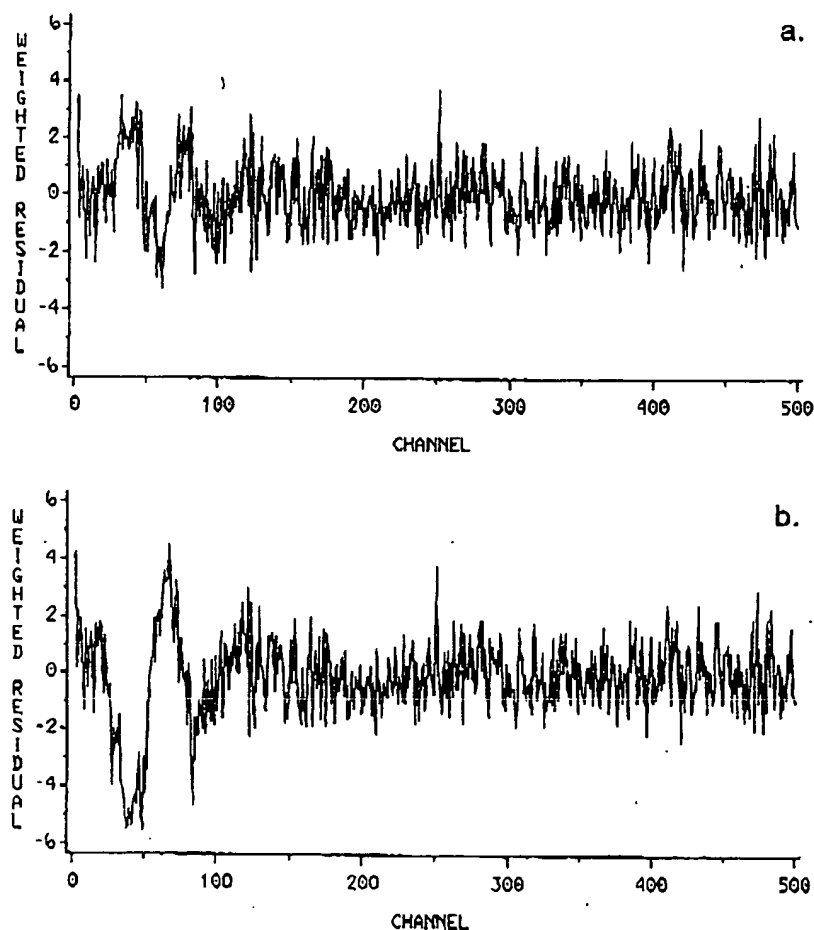


Fig. 2. Effect of errors in the FWHM of the reference decay curve on the weighted residuals. The lifetime parameters are listed in table 4. The FWHM of the simulated sample decay curve was 290 ps. Sample data was analyzed using reference decay curves with FWHM of (a) 285 ps and (b) 295 ps.

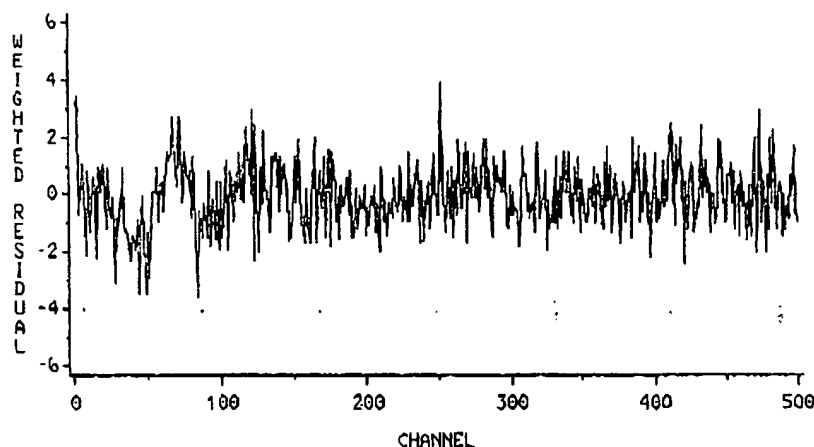


Fig. 3. Effect of a shift in the zero time channel on the weighted residuals. The lifetime parameters are listed in table 4. The reference decay curve was shifted by  $-2.5$  ps with respect to the sample decay curve.

The fitted lifetime parameters are extremely sensitive to variations in the FWHM of the resolution function. The lifetime parameters obtained from an analysis of a three component sample (FWHM = 290 ps) with reference decays having incorrect FWHM values (285 and 295 ps) are given in table 4. The lifetime parameters determined by SPLMOD when the FWHM for the reference is less than that employed for the sample are very poor. The intensities of all components are grossly in error. In addition, a spurious short-lived component appears. When the FWHM of the reference is made larger than that employed for the sample, a spurious long-lived component appears (100 ns). Estimates of the other lifetime parameters are in error, but are not nearly as poor as those determined with the smaller reference FWHM. Plots of the residuals are shown in fig. 2. The fits to the data are poor about the peak.

A shift in the zero channel of the reference with respect to the sample data also generates poor estimates of the lifetime parameters and an additional spurious short-lived component. The example given in table 4 and fig. 3 was obtained by shifting the reference decay curve by  $-2.5$  ps with respect to the sample decay curve. This shift is a fifth of the channel width employed. SPLMOD can correct for small shifts in the zero time channel by assuming that  $y(t + \epsilon)$  has been used instead of  $y(t)$  and then expanding eq. (11) in a Taylor series about  $y(t)$  to the first order term. This gives an additional linear term in eq. (12). However, in this case the correction did little to improve the fit to the data.

The analysis assumes the reference to have a single lifetime component, an ideal which is unlikely to be exact for real systems. Annihilation in the source and trapping at the surface of the reference material undoubtedly introduce additional lifetime components.

The effect of introducing an additional component with a lifetime of 450 ps into the reference decay curve is shown in table 4. The estimated lifetime parameters are relatively insensitive to the presence of the contaminant, even when it represents 15% of the reference decay. The largest error appears in the sample component which has a lifetime closest to the contaminant lifetime.

## 8. Conclusions

The results presented in the previous sections suggest that the deconvolution and least squares analysis methods employed by SPLMOD can provide excellent estimates of positron annihilation lifetime parameters for systems that can be represented by a discrete sum of exponentials. As we have mentioned, this representation is too restrictive for many systems of interest in which distributions of positron lifetimes are expected. For such systems numerical Laplace inversion methods are obviously to be preferred. The results of our tests of a modified version of the program, CONTIN [3,5,6], which solves integral equations with convoluted kernels will be published shortly. It is nevertheless clear from the present work, that estimates of the lifetime parameters are critically dependent on accurate knowledge of the instrument resolution function. We doubt that the deconvolution method employed here is any more sensitive to errors in the resolution function than other algorithms that have been described in the literature or that are currently in use. For example, lifetimes reported using  $^{60}\text{Co}$  prompt spectra to measure the instrument resolution function are undoubtedly in error since it is known that this method provides a poor estimate of the true resolution function occurring with  $^{22}\text{Na}$  sources [3]. There is little doubt that these problems will become

even more severe as we attempt to determine continuous lifetime distributions with numerical Laplace inversion techniques.

#### Acknowledgements

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**DECONVOLUTED DOPPLER BROADENED POSITRON ANNIHILATION  
SPECTROSCOPY: CHARACTERIZATION OF DEFECTS IN ALUMINUM**

*Duke University*

Ph.D. 1985

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Figure 14.	A Comparison of the Peak Region of the Raw Data and the Deconvoluted Data for a Quenched Aluminum Sample.	70
Figure 15.	A Plot of the Error Parameter vs. Quench Temperature.	72
Figure 16.	A Plot of $R_1$ Values for the Raw Data vs. Quench Temperature.	80
Figure 17.	A Plot of $R_1$ Values as Calculated from Average Lineshape Parameter Values for the Raw Data vs. Quench Temperature.	82
Figure 18.	A Plot of $R_1$ Values for the Deconvoluted Data, Using All Retained Coefficients, vs. Quench Temperature.	83
Figure 19.	A Comparison of the Raw Data for an Aluminum Sample Quenched from 600°C and the Same Sample after a High Temperature Anneal.	84
Figure 20.	A Comparison of the Tail Region of the Raw Data for an Aluminum Sample Quenched from 600°C and the Same Sample after a High Temperature Anneal.	85
Figure 21.	A Comparison of the Deconvoluted Data for an Aluminum Sample Quenched from 600°C and the Same Sample After a High Temperature Anneal.	87
Figure 22.	A Comparison of the Tail Region of the Deconvoluted Data for an Aluminum Sample Quenched From 600°C and the Same Sample After a High Temperature Anneal.	88
Figure 23.	A Comparison of the Tail Region of the Same Samples as in Figure 22, but this time Both Data Sets were Deconvoluted Using the Same Number of Coefficients.	92
Figure 24.	A Plot of $R_1$ Values for the Deconvoluted Data Using a Constant Number of Reconstruction Coefficients.	93

## LIST OF FIGURES

	Page
Figure 1. A Schematic Diagram Showing the Effects of the Electron-Positron Pair Momentum on the Energy and Direction of the Two Resulting Annihilation $\gamma$ -Rays in the Laboratory Reference Frame.	9
Figure 2. A Typical Instrument Resolution Function for the Doppler PAS System Used in this Work.	21
Figure 3. A Schematic Diagram of the Quenching Apparatus.	27
Figure 4. A Schematic Diagram Showing the Relationship Between $r(x)$ , $f(x)$ , and $h(x)$ .	30
Figure 5. A Plot Comparing the Power Spectrum of the Annihilation Energy Distribution Before and After Deconvolution.	46
Figure 6. A Semi-Log Plot of the Number of Reconstruction Coefficients Retained by the FFT/PS Algorithm as a Function of the Number of Counts in the Peak.	47
Figure 7. A Comparison of the Raw Data and the Deconvoluted Data for an Irradiated Sapphire Sample.	49
Figure 8. A Comparison of the Raw Data, $h(x)$ , with the Function Obtained by the Convolution of $f(x)$ with $r(x)$ .	50
Figure 9. A Comparison of the Resolution Function Before Deconvolution, After Deconvolution Using 70 Coefficients, and After Deconvolution Using 243 Coefficients.	54
Figure 10. A Plot of the Deconvoluted Lineshape Parameter $x(1)$ as a Function of Quench Temperature.	58
Figure 11. A Plot of the Raw Data Lineshape Parameter $x(1)$ as a Function of Quench Temperature.	62
Figure 12. A Comparison of the Shape of Resolution Functions Collected at Different Times During a Series of Experiments.	64
Figure 13. A Graphical Interpretation of the Sensitivity and Error Lineshape Parameters.	67



F. A LISTING OF THE FORTRAN FFT/PS DECONVOLUTION PROGRAM	117
G. RELATIONSHIP BETWEEN $C_v$ AND $C_d$	141
H. THE UTILITY OF "R" PARAMETER ANALYSIS	143
REFERENCES	145

## TABLE OF CONTENTS

ABSTRACT	(iii)
ACKNOWLEDGEMENTS	(iv)
LIST OF FIGURES	(vii)
CHAPTERS	
I. INTRODUCTION	2
II. EXPERIMENTAL PROCEDURES	17
The Doppler PAS System, 17	
Specimen Preparation, 23	
III. DECONVOLUTION OF DOPPLER PAS MEASUREMENTS	28
The Mathematics of Deconvolution, 29	
Previous Deconvolution Techniques, 33	
The FFT/Power Spectrum Deconvolution Method, 39	
Test of the Method, 44	
IV. RESULTS AND DISCUSSION	52
The Utility of the FFT/PS Deconvolution Algorithm, 52	
The Separation of Multiple Defect Types, 74	
V. CONCLUSIONS AND FUTURE WORK	96
APPENDICES	
A. POSITRON THERMALIZATION TIMES	104
B. THE EQUIVALENCE OF DOPPLER AND ANGULAR CORRELATION MEASUREMENTS	107
C. CHEMICAL ANALYSIS OF THE ALUMINUM	111
D. THE UTILITY OF FOURIER ANALYSIS IN DECONVOLUTION	112
E. THE MATHEMATICAL DIFFICULTIES ASSOCIATED WITH THE INCORRECTLY POSED INTEGRAL EQUATION	114

## ABSTRACT

Positron Annihilation Spectroscopy (PAS) has become a useful tool for the nondestructive investigation of the defect structure of materials. Of the existing PAS measurement techniques Doppler broadening appears to offer several practical advantages. These advantages include low radioactive source strength requirements, comparatively short experimental times, and minimal geometrical restrictions. Unfortunately, the technique suffers from some inherent disadvantages. These include comparatively poor instrumental energy resolution and a limited ability to distinguish between multiple defect types.

In an attempt to overcome the difficulties associated with the use of Doppler broadened PAS the Fast Fourier Transform/Power Spectrum (FFT/PS) deconvolution algorithm has been developed in this work. This algorithm is computationally efficient, noniterative, and requires minimal a priori information. The utility of the FFT/PS deconvolution algorithm is demonstrated for high purity aluminum samples containing known concentrations of vacancies or prismatic dislocation loops. It is shown that the FFT/PS procedure is capable of automatically correcting for accidental and/or intentional changes in the instrument resolution function. In addition, based on the limited data analyzed in this work, it appears that both the sensitivity and the accuracy of the energy distribution lineshape parameters are improved by deconvolution. Finally, it is demonstrated that if the filtering error introduced by the FFT/PS procedure is controlled, then it is possible to distinguish between samples containing predominately vacancy type defects and those with prismatic dislocation loops as the dominant defect type.

DECONVOLUTED DOPPLER BROADENED POSITRON ANNIHILATION  
SPECTROSCOPY: CHARACTERIZATION OF  
DEFECTS IN ALUMINUM

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## CHAPTER I

### INTRODUCTION

Over 50 years ago, Dirac postulated the existence of an atomic particle with a mass equal to that of an electron, but which is positively charged. In 1932 these positively charged electrons, referred to as positrons, were first observed by C. D. Anderson as a by-product of cosmic ray showers. Since that time positron research has been pursued in such varied disciplines as high energy nuclear physics, astrophysics, and materials science. It is within the broad context of materials science, and more specifically in the area of non-destructive testing (NDT) of aluminum, that the interaction of positrons with matter will be examined in this work. Before going on to describe the use of positrons as an NDT tool in the investigation of the defect structure of metals, a brief review of positron physics will be presented. A comprehensive treatment of this subject has been written by West (1974).

As postulated by Dirac in 1935, positrons and electrons are complementary states of matter such that when the two particles encounter one another they will annihilate each other. Their kinetic and rest mass energies will be dissipated as the energy of one, two, or three gamma rays in such a way as to conserve momentum. It can be shown that the ratio of the two-photon to three-photon annihilation event is 372:1 (Stevens 1980), while the ratio of the two-photon to one-photon events is  $\sim 2.6 \times 10^6:1$ , (West 1974). With these ratios as justification the remainder of this work will consider only the annihilation event yielding two photons. If both the electron and positron were at rest, a

positrons in vacancy type lattice defects. This effect was first observed by MacKenzie et al. and Dekhtyar et al. in 1964. Schultz et al. (1981) describe the changes in the annihilation characteristics due to the localization of a positron in a monovacancy type defect as follows: 1) Because the localized positron has a smaller overlap of its wave function with that of the high energy core electrons than it does when diffusing through the nearly perfect lattice, the annihilation rate with high energy core electrons decreases. 2) The density of conduction electrons in the vacancy is also decreased resulting in a decreased annihilation rate with conduction electrons. However, the percentage reduction is less than in the case of core electrons so that the net result is an increase in the fraction of annihilation events involving low energy conduction electrons, thus resulting in a decrease in the width of the Doppler broadened annihilation radiation distribution. 3) The potential for electrons in the vacancy is higher than normal resulting in the conduction electrons traveling more slowly. This in turn also decreases the observed Doppler broadening. 4) A second order effect is that the overlap of the positron wave function with weakly bound outer-core electrons is reduced less than is the overlap with inner-core electrons. This too leads to less broadening of the observed 511 KeV line. 5) The uncertainty principle requires that localization of the positron be accompanied by an increased spread in its momentum distribution (and a corresponding decrease in its de Broglie wavelength). As a result, the contribution of the positron to the center-of-mass velocity increases corresponding to a broadening of the annihilation energy spectrum (Schultz et al. 1981). The first four effects lead to a narrowing of the annihilation energy spectrum while the fifth effect tends to broaden the spectrum. The combination of these effects results in more sharply peaked (narrower) Doppler spectra for metals containing defects which are capable of trapping positrons (Siegel 1980).

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As discussed by Siegel (1980) it is important to note that the positron is not equally sensitive to all lattice defects. It is sensitive to defect regions which can be characterized as having a lower-than-average electron density, for example, vacancies, small vacancy clusters, voids, and dislocation lines or joys. On the other hand, positrons are relatively insensitive to defect regions with a higher-than-average electron density such as interstitials. Indeed this defect specificity is one of the advantages PAS measurements offer as compared with other NDT techniques such as residual electron resistivity (Siegel 1980).

In addition, positrons are capable of probing the entire depth of the valence band (Mackenzie et al. 1964) which is in contrast with techniques such as electrical and thermal conductivity, thermoelectric power, Hall coefficient, magnetic susceptibility, and Knight shift, all of which yield information restricted to the electrons near the Fermi surface.

PAS measurements are an attractive NDT tool since the positron causes negligible perturbation of the electronic structure as it passes through a crystal (Johnson et al. 1978) and the positron-electron interaction does not appreciably alter the momentum distribution of the electrons (Szotek and Chmielewski 1981). A further advantage of the technique is that it yields information regarding both the electron momentum distribution as well as the electron density within the sample (Siegel 1980).

The three most common PAS techniques are lifetime measurements, angular correlation, and Doppler broadening. The effective electron density in the region from which the positron annihilates can most easily be determined using lifetime measurement techniques. These measurements usually use  $^{22}\text{Na}$  as a source material since it simultaneously ( $< 10$  psec) emits a  $\gamma$ -ray along with the positron. This 1.28 MeV  $\gamma$ -ray acts to start the timing apparatus

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which is stopped by the arrival of the  $\sim 511$  KeV  $\gamma$ -ray signifying the occurrence of the annihilation event. The positron lifetime depends on the concentration and distribution of the electrons in the region of the material through which the thermalized positron drifts before its annihilation. This "mean" lifetime will of course be affected by any process, such as the trapping of positrons at defects, which alters the local concentration of electrons seen by the positron (Coleman 1977). These mean lifetimes, which are on the order of hundreds of picoseconds, can be measured to an accuracy of a few picoseconds with modern equipment.

As mentioned previously, when the annihilation event does occur the electron-positron pair will usually have a small net momentum in the laboratory frame. This net momentum affects both the energy of the resulting  $\gamma$ -ray as well as its direction of emission as shown in Figure 1. The relationships between the momentum of the annihilating  $\gamma$ -ray, the measured Doppler shift in energy, and the measured angle  $\delta\phi$  are described in Appendix B.

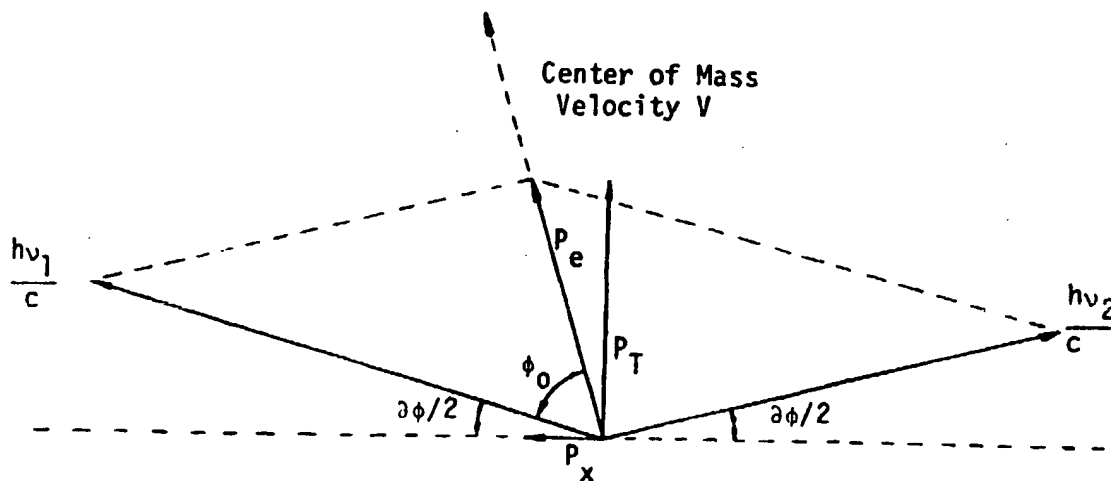


Figure 1. A Schematic Diagram Showing the Effects of the Electron-Positron Pair Momentum on the Energy and Direction of the Two Resulting Annihilation  $\gamma$ -Rays in the Laboratory Reference Frame.

Angular correlation measurements, which measure the deviation from colinearity of the two photons emitted from the annihilation process, were first reported by DeBenedetti and his coworkers in 1950. The so called long-slit geometry (DeBenedetti 1950) is commonly chosen since it provides both good angular resolution and adequate counting rates. However, other geometries can be employed for specific applications. The angular distribution of a sample is determined by measuring the coincidence counting rate as a function of angular deviation from colinearity. In well annealed metals this data generally takes the form of a central inverted parabola with Gaussian wings. The instrument resolution is good, typically  $\sim 0.5$  mrad, but the counting rate is comparatively slow and the required source strengths are relatively high.

Alternatively, Doppler broadening measurements could be used to determine the net momentum distribution of the annihilating electron-positron pairs by measuring the energies of the resulting  $\gamma$ -rays. The center of mass momentum of the annihilating pair along the line of emission of the  $\gamma$ -ray gives rise to Doppler shifts of the  $\gamma$ -ray energies. The distribution of these momentum components then results in a Doppler broadening to give the 511 KeV line a width of approximately  $511 (V_F/2c)$  KeV where  $V_F (\sim 10^8 \text{ cm/s})$  is the Fermi velocity of the conduction electrons and  $c$  is the speed of light (Hughes 1982). Details of the experimental procedure for collecting Doppler-broadened spectra are given in Chapter II.

Although lifetime measurements, angular correlation, and Doppler-broadening measurements are by far the most common PAS techniques there are two others which should be mentioned. One of the drawbacks of using conventional sources such as  $^{22}\text{Na}$  or  $^{68}\text{Ge}$  is that only certain depths of the material under investigation are sampled extensively by these positrons. Hughes et al. (1982) give the equation:

$$\delta E = \pm \frac{1}{2} M_0 c^2 (\delta\theta) = \pm 255.5 (\delta\theta) \text{ eV} \quad (1.2)$$

where  $\delta E$  is the Doppler Energy Resolution in eV  
and  $\delta\theta$  is the Angular Correlation Resolution in mrad.

The best Doppler systems have a resolution of about 1.2 KeV which corresponds to an angular correlation resolution of 5 mrad. Typical angular correlation systems have a resolution approximately an order of magnitude better than this. For Doppler broadening techniques to effectively compete with angular correlation techniques in high precision applications it is necessary to improve the effective resolution of Doppler PAS. Schultz et al. (1981) point out that theoretically a 10-fold increase in effective resolving power of Doppler measurements can be achieved by deconvolution.

Deconvolution is the mathematical technique used to separate the instrumental broadening effects from the true Doppler broadened positron annihilation  $\gamma$ -ray spectrum. Instrumental broadening is a complicated problem caused by many different factors. For example, physical properties of the detector crystal such as frontal dead layers, geometry of the active region, and regions of poor charge collection, can all lead to line broadening. The primary contribution to instrumental broadening in a solid state semiconductor detector is due to statistical fluctuations in the division of absorbed energy between ionization and heating of the crystal lattice, i.e. the Fanno effect, (McNelles and Campbell 1975).

The shape of the instrument resolution function, as well as the magnitude of the line broadening, is most often determined from the 514 KeV  $\gamma$ -ray emitted from the isotope  $^{85}\text{Sr}$ . The difficulty lies in the fact that the Doppler-broadening of the 511 KeV  $\gamma$ -ray line in the absence of defects in a metallic crystal is of the same order ( $\sim$  KeV) as the detector resolution, and

changes induced by defects are significantly smaller. Fortunately, the defect induced changes in the deconvoluted  $\gamma$ -ray energy distribution are much more dramatic than in the instrumentally smeared raw data (McNelles and Campbell 1975). In addition, removing instrumental effects offers another advantage. Such a procedure allows direct comparison of experimental results obtained in different laboratories. If results are reported without deconvolution they must be nondimensionalized in some way or else the instrument resolution function must also be reported. In either case the interpretation of results is complicated in the absence of a deconvolution procedure.

In light of the above discussion it was decided that the results presented in this work would be for deconvoluted Doppler spectra. Unfortunately, the mathematics involved are somewhat complicated. Because of this, most of the existing deconvolution schemes are iterative in nature and hence computationally inefficient. One of the major goals of this work was to develop a deconvolution scheme for Doppler broadened positron annihilation measurements which is model independent, does not require iteration, and which yields reproducible results.

In addition to the need for deconvolution, DBPAS also currently suffers from an inability to distinguish various defect types. For example, vacancies and dislocations tend to alter the annihilation lineshape in qualitatively the same manner. That is, in both cases the spectrum becomes more narrow as the defect concentration increases. Most of the lineshape parameters currently used to characterize Doppler broadened spectra are unable to distinguish the effects of different defect types because they describe only gross changes in lineshape. For example, MacKenzie's S-parameter, which is the most commonly used lineshape parameter, essentially measures the width of the distribution (MacKenzie et al. 1970). Hence, for any given spectrum such

an analysis yields a single value. This is useful if there is only one degree of freedom in the series of experiments. If two variables are allowed to change, however, it is impossible to separate their effects. Clearly, additional parameters are required. The other parameters selected must be uncorrelated with the S-parameter. That is, if the second parameter chosen is a function of the first parameter only, then no new information is gained. Campbell (1977) and Schultz et al. (1981) have discussed the use of other possible parameters to describe lineshapes. Although these parameters may be more sensitive to variations in specific defect concentrations they are correlated with S, and therefore of little help in differentiating defect types.

In light of the above discussion it can be seen that it would be desirable to find a family of lineshape parameters which is capable of distinguishing various types of metallurgical defects. For example, one of the groups of parameters to be examined in this work is the central moments of the  $\gamma$ -ray energy distribution. It is well known that the values of these moments give information regarding the shape of the distribution. In fact, it can be shown that if all of the moments are known one can construct the distribution. The even moments give information about the width of the distribution while the odd moments vanish for symmetrical distributions such as the ones to be examined in this work (the symmetry question is addressed in Chapter III). The second central moment is called the variance and its square root is the familiar standard deviation. The fourth central moment is called the Kurtosis or flatness. The advantage of moment analysis is that the information obtained is not redundant. For example, it is very possible to have two distributions with equal standard deviations (second moments) and yet widely different fourth moments. Such a family of lineshape parameters suggests the

---

possibility of separating the effects of multiple metallurgical defect types using DBPAS.

The above brief review of positron annihilation spectroscopy as an NDT tool for the study of metallurgical defects indicates that the technique with the greatest potential for practical use outside the laboratory is Doppler-broadening. This technique offers the advantages of low source strength requirements, the ability to use  $^{68}\text{Ge}$  as a source material in order to reduce surface effects, and few geometrical experimental constraints. Doppler broadening currently suffers from relatively poor energy resolution and an inability to distinguish different metallurgical defect types. Based on these observations, the purpose of this work is twofold: 1) to develop an efficient, model independent, noniterative deconvolution method, and 2) to develop a family of lineshape parameters capable of distinguishing various defect types.

## CHAPTER II

### EXPERIMENTAL PROCEDURES

This chapter is divided into two sections. The first section describes the Doppler broadened PAS instrumentation and the methodology employed to collect Doppler spectra. The second section outlines the thermomechanical treatments used to create the desired defect concentrations in the metallic samples which were studied in this work.

#### The Doppler PAS System

The heart of the Doppler broadened PAS instrumentation system is the single crystal high purity Germanium (HPGe) detector. This detector is mounted at the end of a cold finger in a cryostat and, to reduce thermal noise, is operated at liquid nitrogen temperatures. The cylindrical detector crystal used in this work is 46.7 mm in diameter and 46.2 mm in length. When the annihilation  $\gamma$ -rays impinge upon the crystal their energy is absorbed by two mechanisms. The first is the creation of electron-hole pairs within the crystal. These electron-hole pairs are subsequently swept apart by the applied detector bias (4000 V) and flow through an external circuit creating a current pulse whose integral with respect to time has an amplitude proportional to the incident  $\gamma$ -ray energy. The second absorption mechanism is heating of the crystal. The ratio of these two absorption mechanisms is known as

the Fanno factor and the statistical fluctuations in the division of absorbed energy between ionization and heating is the primary contribution to instrumental line broadening in solid state semiconductor detectors. For further details of the absorption process see Moore (1983).

The resulting current pulse from the detector is rather weak and unsuitable for cable transmission. Because of this a preamplifier (Ortec Model 120-5) is mounted directly adjacent to the detector output. The detector and preamp are D.C. coupled, as is the entire system, and powered from the same A.C. source to prevent ground loops. The preamp is a charge sensitive device capable of accepting signals corresponding to the  $\gamma$ -ray energy range of 0-10 MeV. Its output is a voltage pulse with an amplitude proportional to the integrated charge supplied by the detector and hence proportional to the incident  $\gamma$ -ray energy. The magnitude of the voltage pulse amplitude is related to the incident  $\gamma$ -ray energy by the approximate conversion factor of 280 mV per MeV. Therefore for a typical 511 KeV  $\gamma$ -ray the output of the preamp would be approximately 143 mV. In addition to converting the detector charge pulse to a voltage pulse with an intensity suitable for cable transmission the preamp also shapes the resulting voltage pulse so that it is acceptable to the linear amplifier.

The linear amplifier used in this work is an Ortec Model 571. It accepts the shaped pulses from the preamp and amplifies them to suitable voltages for transmission to the multichannel analyzer. The output pulses are active filter shaped (skewed Gaussian approximations) and have amplitudes in the 0-10 V range. Unipolar outputs are used to optimize the signal-to-noise ratio (resolution). Since the main amplifier is operated within its linear range the amplitude of the output voltage pulse is proportional to the incident  $\gamma$ -ray energy as measured by the HPGe detector.

---



The output from the linear amplifier is routed to an Ortec Model 408A biased amplifier. This device accepts the output pulses from the main amplifier and allows one to selectively view any region of the initial 0-10 MeV energy range. In this work the biased amplifier was used to select the energy window of 483 to 548 KeV. The output pulses from the biased amplifier serve as the input to the analog-to-digital (ADC) converter located within the Norland IT-5300 multi-channel analyzer (MCA). In this device the amplitude of the voltage pulse is converted to a digital representation of the energy of the detected  $\gamma$ -ray and subsequently dynamically displayed on the MCA's CRT in the form of a histogram of energy versus count intensity. The MCA can be programmed to accept input signals until a predetermined number of counts in the peak channel is reached. At this time the experiment is terminated and a complete Doppler broadened spectrum is displayed on the CRT. The energy distribution is then ready for transmission to the computer for subsequent data manipulation.

The two most important parameters which characterize the quality of a Doppler PAS system are the width, in units of energy, of the instrument resolution function and the energy resolution, in units of eV per channel, of the MCA. The width of the instrument resolution function (IRF) is, in essence, a measure of the quality of the detector crystal, although it is also affected by the associated amplification electronics as well as the ADC. The IRF is most often determined by examining the response of the system to the monoenergetic ( $E = 514$  KeV) photons emitted from the isotope  $^{85}\text{Sr}$ . If the instrumentation was perfect the energy distribution, as recorded by the MCA, would consist of a single bar in the histogram in the channel corresponding to 514 KeV. However, in any real instrumentation this line will be broadened for reasons previously discussed. The amount of instrumental broadening is most often re-

ported in terms of the energy width of the  $^{85}\text{Sr}$  line as measured across the two channels which contain half the number of counts in the peak channel. This energy measure is commonly named the full width half maximum (FWHM). Figure 2 shows typical IRF for the Doppler PAS system used in this work. As can be seen in this figure the FWHM of the IRF is about 1.28 KeV.

The second parameter which describes the quality of the Doppler PAS instrumentation is the energy resolution of the MCA which is determined by the following procedure. First, a Doppler broadened spectrum is collected and the channel corresponding to its centroid is determined. The centroid channel is assigned the energy value 511 KeV. Next, an energy spectrum is obtained for the isotope  $^{85}\text{Sr}$ . The channel corresponding to the centroid of this distribution is calculated and assigned the energy value 514 KeV. The MCA energy resolution is then easily calculated by dividing the energy difference (3 KeV) by the number of channels between the centroids. In this work the 511 KeV channel was number 439 while channel number 486 corresponded to 514 KeV. Hence, the MCA energy resolution is  $3000 \text{ eV}/(486 - 439)$  or  $\sim 64 \text{ eV per channel}$ . Clearly this value can be adjusted using the biased amplifier. The reason for selecting the 64 eV/channel is to obtain the best possible resolution while maintaining the ability to display the entire region of interest (483 to 548 KeV) on the 1024 channel MCA. The reasons for requiring the above energy window will be discussed in Chapter III.

While the MCA energy resolution is a parameter which, to a certain extent, can be varied, the IRF is essentially predetermined by the instrumentation. The one factor contributing to the width of the IRF which can be controlled is the data acquisition, or count rate. It is well known that increasing the count rate decreases the instrument resolution. Therefore, one must select a count rate which results in an acceptable line broadening while

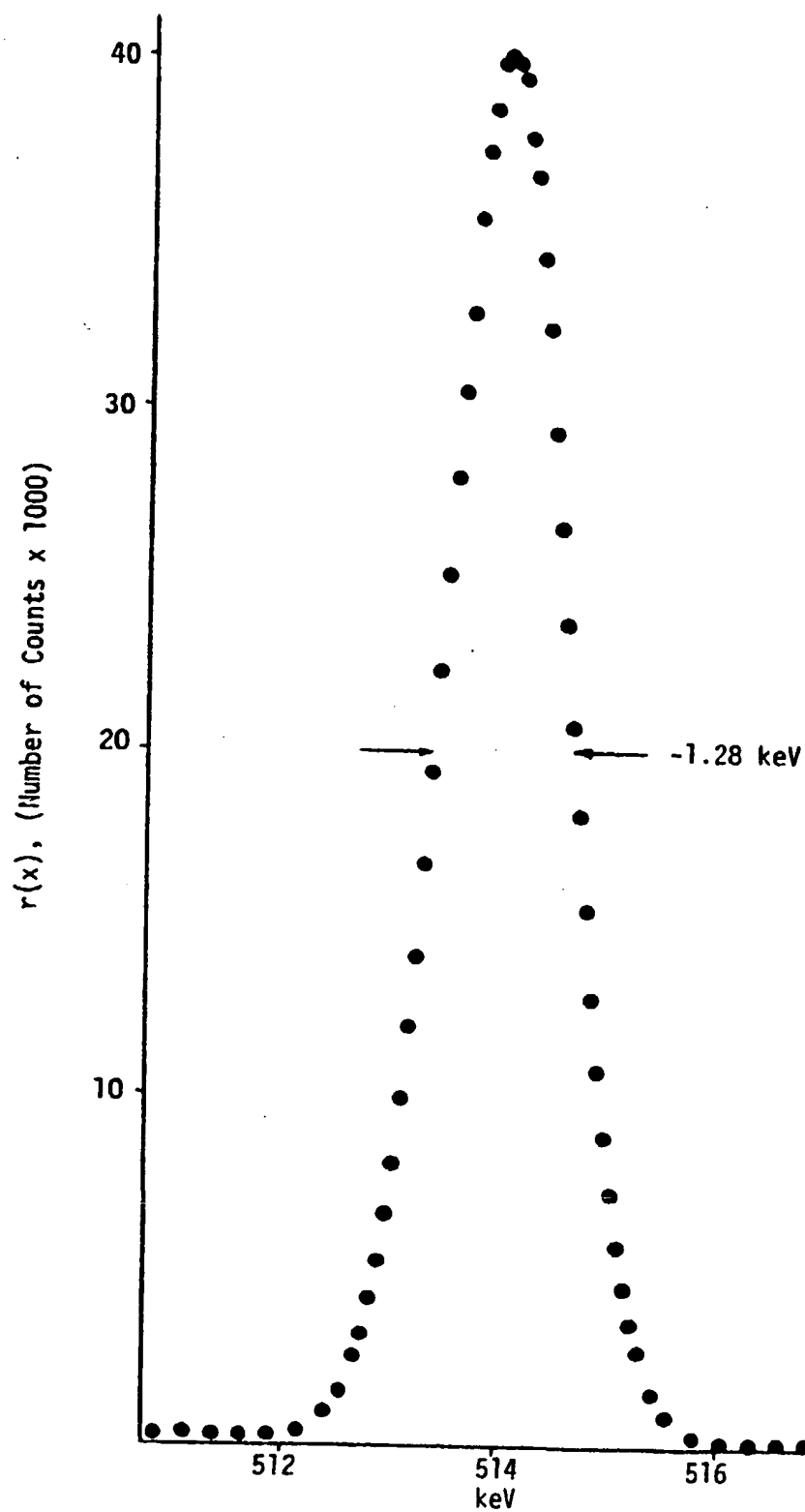


Figure 2. A Typical Instrument Resolution Function for the Doppler Pas System used in this Work. Note that the Full Width Half Maximum is about 1.28 keV.

keeping the time required to collect a spectrum reasonably short. Moore (1983) demonstrated that with the Doppler instrumentation used in this work very reproducible results can be obtained with count rates of  $\leq 2$  kHz. The count rate is easily adjusted by changing the sample to detector distance. All of the results reported in this work were obtained using count rates of  $\leq 2$  kHz with an associated experiment time of  $\sim 30$  minutes resulting in a peak height of 40K counts and  $\sim 3 \times 10^6$  total integrated counts.

The positron source isotope in all experiments was  $^{68}\text{Ge}$  which was chosen to avoid surface effects. The mean depth of penetration in aluminum for such positrons is  $\sim 0.50$  mm. The activity of the source was initially 50  $\mu\text{C}$ . The half-life of  $^{68}\text{Ge}$  is  $\sim 275$  days and hence the activity of the source did not change significantly for any single set of experiments since a typical set of measurements requires less than a week to accumulate. It should be pointed out that the activity of the source does not measurably affect the observed annihilation spectra, provided the count rate remains constant.

Although the count rate can be easily adjusted by changing the source to sample distance such a change introduces an additional error source. This error source is related to the source/sample geometry. It has been demonstrated that the observed spectra may depend on the angle with which the gamma rays impinge upon the detector crystal (Smedskjaer 1983). It has also been shown that in high precision experiments among samples of the same material, one should use samples of nearly identical thickness to reduce the error associated with small angle scattering (Smedskjaer 1983).

The  $^{68}\text{Ge}$  source was deposited on a thin ( $\sim 0.05$  mm) nickel foil for handling convenience. This results in what is known as a source contribution to the annihilation lineshape. That is, in each experiment some of the positrons will annihilate in the nickel foil and the  $\gamma$ -rays produced by these

events will contribute to the overall annihilation spectrum. However, the ratio of source annihilation events to sample annihilation events is small ( $< 1\%$ ), and the source contribution is neglected in the remainder of this work.

In all measurements the source and sample were arranged in what is commonly called the sandwich configuration. In this configuration the positron source is placed between two identically treated metallic samples. The samples are thick enough, typically a few millimeters, to guarantee that virtually all of the positrons annihilate within the material being examined.

### Specimen Preparation

The material examined in this work is high purity, large grain size aluminum purchased from the Materials Research Corporation. The aluminum was zone refined to a nominal purity of 99.9995% before being vacuum melted and cold fabricated to size. The chemical analysis, in PPM of metallic interstitials, is listed in Appendix C. The aluminum was supplied as a 13 mm diameter rod from which  $\sim 1.5$  mm thick disks were machined. Each of these disks had an  $\sim 1.5$  mm hole drilled through it near its edge. The purpose of this hole was to allow the disks to be suspended from a titanium wire while within the vertical tube furnace. After the disks were machined to size, they were given a full anneal heat treatment consisting of 2 hours at  $400^{\circ}\text{C}$  followed by furnace cooling overnight. The as annealed aluminum disks were the starting material for all subsequent thermal treatments.

The thermal treatments used in this work are very similar to those outlined by Cotterill et al. (1972). In this work the authors describe a two step time/temperature cycle which allows one to produce, in a controlled fashion, first samples with vacancies as the dominate defect type, and then,

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samples with dislocation loops as the major defect type. The actual thermal treatment employed in this work is as follows:

- 1) Heat the samples to the desired quenching temperature and hold them at temperature for one hour to establish thermal equilibrium.
- 2) Quench the samples from the furnace into  $\sim 293^\circ\text{K}$  water.
- 3) Dry the samples within 5 to 10 seconds.
- 4) Quench from  $\sim 293^\circ\text{K}$  to a liquid nitrogen bath at  $77^\circ\text{K}$ . This procedure has been shown to produce primarily vacancies, the concentration of which is a direct function of the quenching temperature.
- 5) After DBPAS characterization, the aluminum disks are then annealed at  $353^\circ\text{K}$  for thirty minutes, which results in the production of prismatic dislocation loops by vacancy coalescence.
- 6) The samples are cooled to  $77^\circ\text{K}$  and the PAS measurements corresponding to the dislocation loops are made.
- 7) Finally the disks are annealed at  $523^\circ\text{K}$  for two hours to remove the dislocations and return the material to the annealed state.

Several of the steps in this procedure require further explanation. The quenching procedure is taken from Cotterill et al. (1972) and their results together with the following discussion forms the basis for the assumption used in this work that the PAS measurements made after step 4 are for a fairly uniform distribution of vacancies of concentration approximately equal to the equilibrium concentration at the quenching temperature.

Quenching experiments are usually performed on thin metallic foils so that the quench rate remains constant across the sample. However, as discussed previously, constraints imposed by the positron annihilation experiments put a lower limit on the sample thickness of  $\sim 1.5$  mm. For a complete review of the complications which can evolve during quenching due to increased

sample thickness see, for example, Cotterill et al. (1965). The main problems to be addressed here are the formation of vacancy clusters and the loss of vacancies at the sample surfaces. The second problem was investigated in a paper by Wilkes and Harrison (1972) in which they observed the quenching of 3 mm thick disks of aluminum and reported a very low vacancy loss. Since the samples used in this work are only  $\sim 1.5$  mm thick and since the quench procedure is similar to that employed by Wilkes and Harrison, it will be assumed that the vacancy loss during the quench can be ignored. As for the formation of vacancy clusters, the positron lifetime measurements of Cotterill et al. (1972) demonstrate that for this quenching procedure the probability of forming bivacancies, or larger clusters, is small.

It is very important to dry the samples thoroughly between the water quench and the liquid nitrogen quench. This is necessary to avoid the formation of a thin layer of ice on the specimen surface which would contribute an undesirable component to the measured PAS spectrum. As pointed out by Cotterill et al. (1972) the annealing of the vacancies during this short drying procedure may be neglected since the mean lifetime of vacancies (at room temperature) in a specimen quenched from 873°K is  $\sim 100$ s and increases for lower quenching temperatures. It is, of course, desirable to keep the drying time as short as possible.

Step 5 of the procedure, the formation of prismatic dislocation loops from the collapse of disk shaped vacancy clusters during a low temperature anneal of aluminum, was reported by Hirsch et al. in 1958. Cotterill et al. (1972) demonstrate that the appropriate thermal treatment consisted of a 30 minute anneal at 353°K. The theoretical relationship between the vacancy concentration after the quench,  $C_v$ , and the concentration of dislocation cores,  $C_d$  (given as an atomic fraction), is presented in Appendix G.

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The apparatus used to perform the thermal treatments is shown in Figure 3. A quartz tube passes through a vertical tube furnace and is attached to the room temperature water reservoir by a "quick disconnect" coupling. Just above the water level in the reservoir is the argon supply port. All thermal treatments were performed in an argon atmosphere to avoid oxidation. The other end of the tube is connected to a flow meter which is used to monitor the argon flow rate. The other side of the flow meter is vented to atmosphere. The temperature is measured by a chromel-alumel thermocouple positioned inside the tube furnace and adjacent to the aluminum disks. These disks are supported by a titanium wire, the other end of which is connected to an iron slug. The slug, and hence the specimens, are held in position in the furnace by a magnet located outside of the tube. The actual quench is performed simply by pulling the magnet away from the tube and allowing the entire specimen-wire-slug arrangement to fall by gravity into the quench bath. It is, of course, important for the system to be designed so that the specimens fall freely through the tube and quickly reach the quench bath. If the specimen-wire-slug arrangement does not fall freely the temperature of the samples when they enter the water will be significantly lower than the temperature measured by the thermocouple.



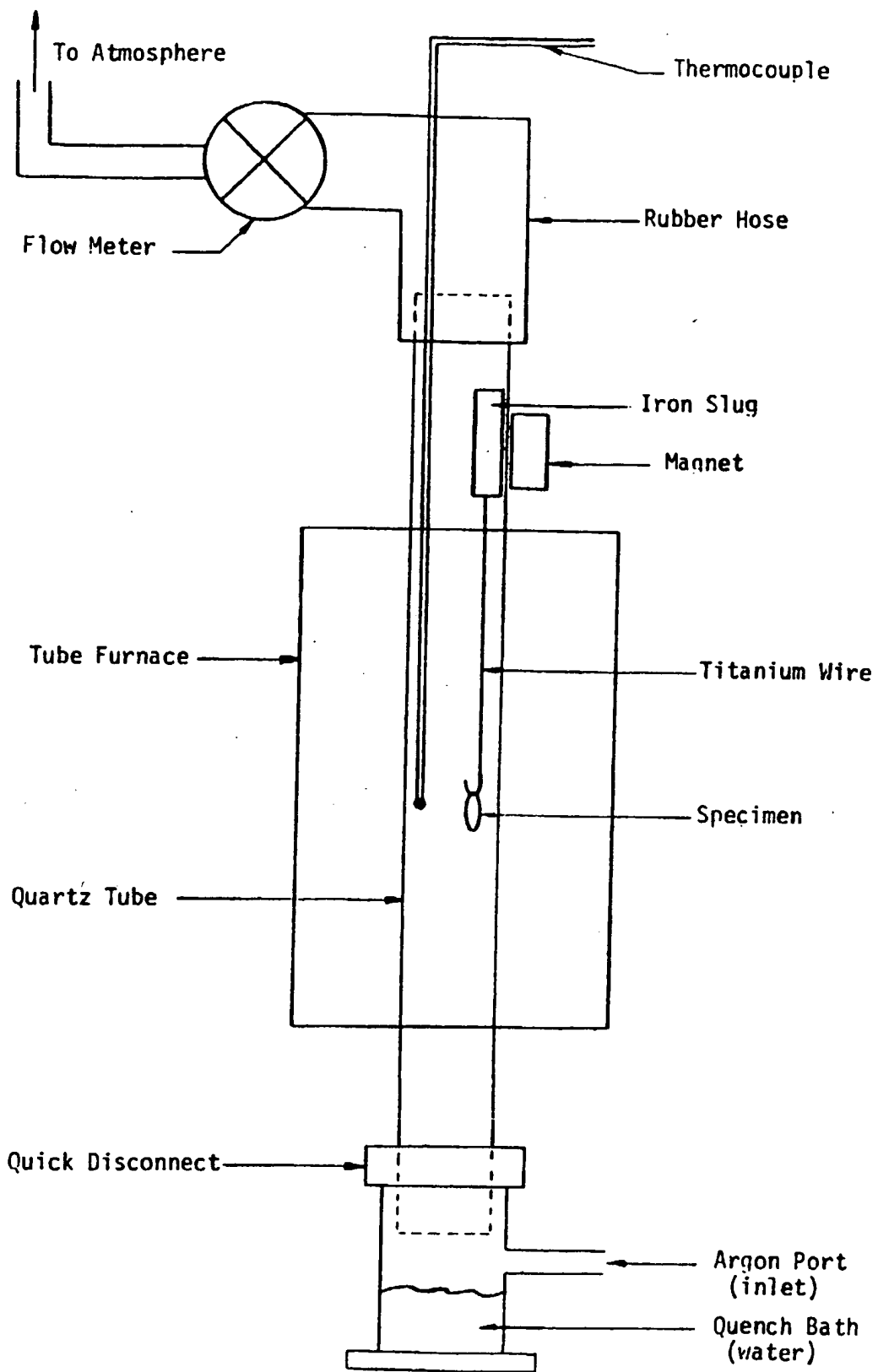


Figure 3. A Schematic Diagram of the Quenching Apparatus.

### CHAPTER III

#### DECONVOLUTION OF DOPPLER PAS MEASUREMENTS

As discussed in the introduction, compared to angular correlation (AC) measurements, the major disadvantage of the Doppler PAS technique is its inherently poor energy resolution. In order for Doppler methods to be competitive with AC methods in high resolution applications the instrumental line broadening must be removed from the experimentally obtained Doppler spectra. Mathematically, the technique which accomplishes this separation is deconvolution. In this chapter the deconvolution of Doppler PAS measurements will be discussed.

The chapter is divided into four sections. In the first section the mathematics of deconvolution are presented. This section justifies the use of deconvolution techniques to separate instrumental effects from the desired true Doppler-broadened energy spectrum. In addition, the inherent advantages and mathematical difficulties associated with the use of Fourier analysis to solve the problem are addressed.

In the second section of this chapter the methods introduced by other investigators to solve the incorrectly posed deconvolution problem, along with their inherent advantages and disadvantages, are discussed. The third section presents the FFT/power spectrum deconvolution method (Schaffer et al. 1984); and describes its use in detail. In the final section of this chapter the FFT/power spectrum method is demonstrated using a neutron irradiated high purity single crystal sapphire sample.

### The Mathematics of Deconvolution

The first objective of this section is to demonstrate the validity of using the convolution equation to describe the relationship between  $h(x)$ , the experimentally observed energy distribution,  $f(x)$ , the true Doppler broadened energy distribution, and  $r(x)$ , the instrument resolution function (IRF). In Figure 4(a) the instrument resolution function  $r(x)$  is shown. Recall that this function represents the instrumental broadening of what should be a single energy line at the centroid of  $r(x)$ . The function  $r(x)$  is assumed to be normalized to unit area. In order to understand the relationship between these three functions,  $f(x)$  should be viewed as a histogram comprised of segments of width  $\Delta x$ . If it can be assumed that the instrumental response is linear (the detector and all associated electronics are operated within their linear range) then each segment of the  $f(x)$  histogram will be broadened by the unit area IRF  $r(x)$ . Hence, the value of the observed function  $h(x)$  at  $x'$  will have contributions from many of the  $f(x)$  histogram segments in the neighborhood of  $x'$ . Figure 4(b) is a schematic representation of the contribution to  $h(x')$  from the histogram section of  $f(x)$  at a distance  $x_1$  from the origin. This contribution  $\Delta h_1(x')$  will be the product of the area  $f(x_1)\Delta x$  and  $r(x' - x_1)$ , that is:

$$\Delta h_1(x') = f(x_1)r(x' - x_1)\Delta x \quad (3.1)$$

The total value of  $h(x')$  will be the sum of the contributions from all possible histogram segments in  $f(x)$ . Thus,

$$h(x') = \sum_{i=-\infty}^{\infty} \Delta h_i(x') = \sum_{i=-\infty}^{\infty} f(x_i)r(x' - x_i)\Delta x \quad (3.2)$$

or as the width of the segments approaches zero the summation in Eq. 3.2 is replaced by an integral to yield,

$$h(x') = \int_{-\infty}^{\infty} f(x)r(x' - x)dx \quad (3.3)$$

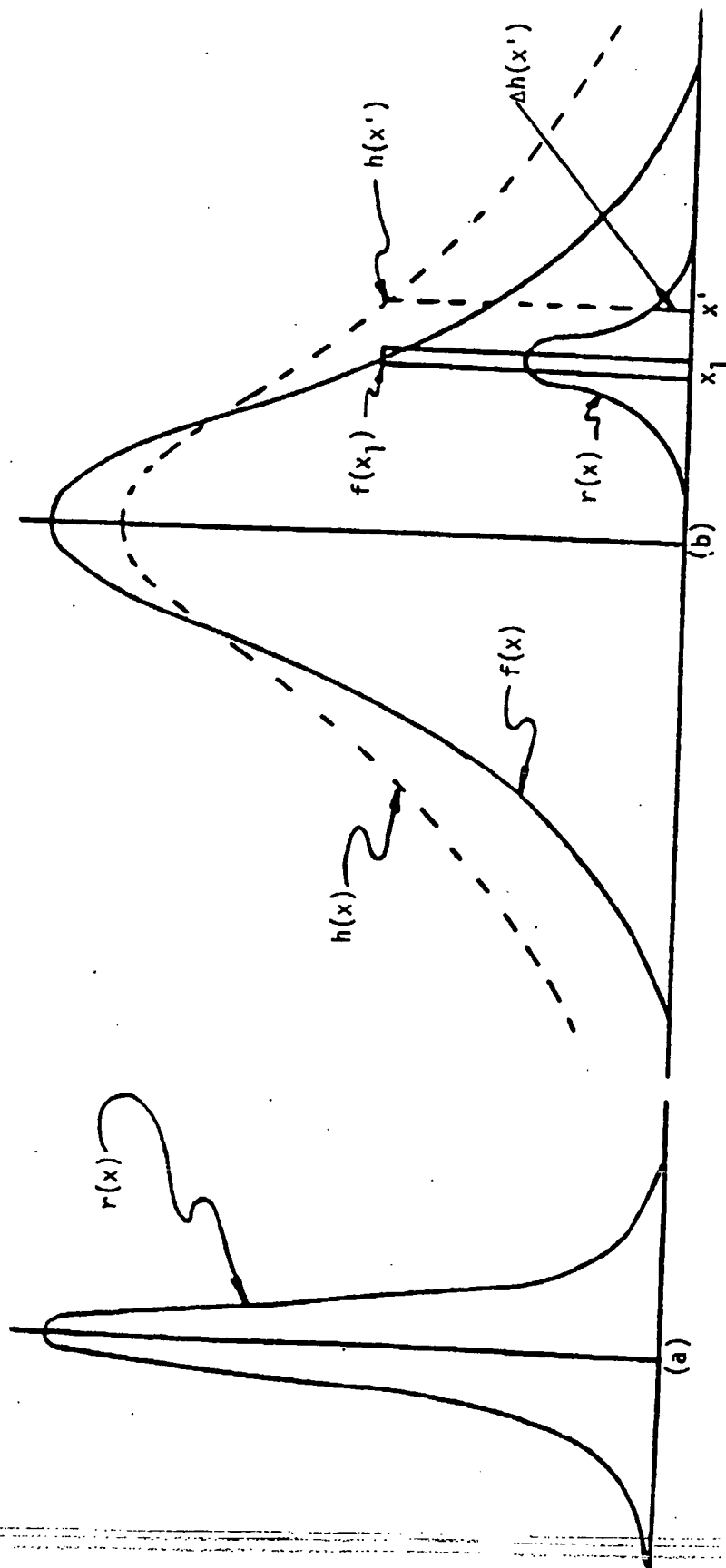


Figure 4. (a) The Instrument Resolution Function  $r(x)$ , (b) A Schematic Representation of the Contribution to  $h(x')$  from the Histogram Section of  $f(x)$  at a Distance  $x_1$  from the Centroid.

Clearly the same procedure holds for any value of  $x$  in  $h(x)$  and the final form of the relationship, with a change of variables, is:

$$h(x) = \int_{-\infty}^{\infty} f(y)r(x - y)dy \quad (3.4)$$

Eq. 3.4 is the convolution equation which serves as the starting point for the remainder of the mathematics used in this work.

The advantage of using Fourier analysis when dealing with convolution problems can be seen clearly when Eq. 3.4 is rewritten as:

$$H(w) = F(w) \cdot R(w) \quad (3.5)$$

where the functions in Eq. 3.5 are the Fourier transforms of the respective functions in Eq. 3.4 according to the general relationship:

$$G(w) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-iwx} g(x) dx \quad (3.6)$$

Note that in Fourier space convolution is simply a complex multiplication. The equivalence of Eqs. 3.4 and 3.5 is demonstrated in Appendix D.

Rearranging Eq. 3.5 yields

$$F(w) = H(w)/R(w) \quad (3.7)$$

from which one can find  $f(x)$  using the inverse Fourier transform

$$f(x) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{iwx} \frac{H(w)}{R(w)} dw \quad (3.8)$$

Eq. 3.8 is an expression for the true Doppler broadened energy spectrum in terms of the Fourier transforms of the experimentally observed function  $h(x)$  and the instrument resolution function  $r(x)$ . However, the uniqueness of the solution given in Eq. 3.8 requires that  $h(x)$  be known exactly. This condition is never satisfied in a real experiment and this fact significantly complicates the mathematics of the problem. The observed function  $h^*(x)$  will be related to the "true" function  $h(x)$ , as defined in Eq. 3.4, for an unknown

function  $f(x)$  by:

$$\max_{-\infty < y < +\infty} |h^*(x) - h(x)| < \epsilon \quad (3.9)$$

where  $\epsilon$  is a measure of the noise associated with the stochastic nature of the experiment.

It can then be shown (see Appendix E) that there are two solutions  $f_1(x)$  and  $f_2(x) = f_1(x) + C \sin(2\pi nx/(b-a))$  with corresponding function  $h_1$  and  $h_2$  (as defined by Eq. 3.4) both of which satisfy the inequality (3.9). Hence, both  $f_1$  and  $f_2$  must be considered as solutions to Eq. 3.4 with a left hand side  $h^*(x)$  taken from an experiment to within an error  $\epsilon$  (Turchin et al. 1971). In fact, there is an infinite set of such solutions a subset of which contain indefinitely large rapidly oscillating components.

In order to understand the mathematical source of these difficulties Eq. 3.8 was investigated further. If in Eq. 3.8  $H(w)$  is replaced by  $H^*(w)$

with

$$H^*(w) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{iwx} h^*(x) dx \quad (3.10)$$

and

$$h^*(x) = h(x) + \delta(x) \quad (3.11)$$

where  $\delta$  is a stationary random process with zero mean, then it has been shown (Rautian 1958) that the dispersion of  $f(x)$  is

$$D(f) = \langle f^2 \rangle - \langle f \rangle^2 = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \frac{G(w)}{|R(w)|^2} dw \quad (3.12)$$

where  $G(w)$  is the power spectrum of  $\delta(x)$

and  $|R(w)| \rightarrow 0$  for  $|w| \rightarrow \infty$

Therefore the dispersion of the solution  $f(x)$  can remain finite only if the power spectrum of the noise approaches zero rapidly as  $w$  approaches infinity. In practice this will not be the case since the noise will inevitably contain a "white noise" component such that  $G(w)$  remains finite as  $|w| \rightarrow \infty$  and the dis-

persion of the solution  $f(x)$  therefore becomes infinite (Turchin et al. 1971).

Alternatively one can understand the source of the mathematical difficulties as follows. The observed function  $h^*(x)$  contains high frequency noise components which are not physically realizable in the true function  $h(x)$ . In Eq. 3.8 these high frequency components are divided by small eigenvalues,  $R(w)$ , which results in large oscillations in the solution  $f(x)$  (Turchin et al. 1971).

Recognizing these difficulties, previous investigators have proposed several deconvolution techniques which appear to successfully solve the problem. Since the accuracy of the deconvoluted spectrum can never really be determined, except in certain carefully constructed numerical experiments, the criticisms of a specific method tend to relate to the amount of computer time required, the complexity of the programming effort, the degree to which the analysis may be automated, or to the need for some a priori information concerning the function  $f(x)$ . As a preface to the introduction of the FFT/power spectrum deconvolution technique, the following section is a review of the prior techniques that have been developed to deconvolute Doppler PAS data.

### Previous Deconvolution Techniques

One of the early Doppler PAS deconvolution schemes introduced by Hotz et al. (1968) is basically a matrix approach. The convolution in its discrete form is

$$h(x) = \sum_{y=-a}^a r(y - x)f(y)\Delta y \quad (3.13)$$

where  $\Delta y$  is the incremental width (resolution)

and the other functions have their usual meanings

This can be rewritten in matrix form as

$$b_i = A_{ij}c_j \quad (3.14)$$

where  $b$  is the discrete experimental vector (spectrum)

$c$  is the unknown discrete vector before instrument broadening

and  $A$  is a rectangular matrix in which each row is the detector response function appropriate to the corresponding component of  $c$ .

In the method developed by Hotz et al. (1968) the matrix  $A$  is determined using  $^{85}\text{Sr}$  and the vector  $b$  is the output of the Doppler system MCA for a given experiment. In this technique an initial estimate of the unknown vector  $c$  is made, say  $c^{(1)}$ , and a corresponding vector  $b^{(1)}$  is calculated from Eq. 3.14. The quality of  $c^{(1)}$  is determined by finding  $\chi^2$  for the fit between  $b$  and  $b^{(1)}$  as defined by

$$\chi^2 = \sum_i (b_i^{(1)} - b_i)^2 / b_i \quad (3.15)$$

Next  $c^{(1)}$  is modified in a manner described in the original work to create a second estimate,  $c^{(2)}$ , of the desired function  $c$ . Then  $b^{(2)}$  is found from Eq. 3.14 and a new  $\chi^2$  value determined. The process is repeated until  $\chi^2$  is minimized. This process is extremely involved and inefficient. In addition the initial estimate  $c^{(1)}$  as well as the modification criteria used to generate subsequent  $c^{(i)}$  estimates can influence the final result.

In more recent years the matrix approach has been examined in greater detail (Aulick and Gallie 1981). Although this work is not related to Doppler PAS measurements, the mathematical problem is the same. It is demonstrated that 3.14 should be rewritten as:

$$\hat{b} = Ac \quad (3.16)$$

where  $\hat{b} = b + \epsilon$

and  $\epsilon$  is the measurement error

The least squares solution of Eq. 3.16 is then



$$A^t \hat{b} = c \quad (3.17)$$

where  $A^t$  is the pseudo-inverse (Penrose 1956) of  $A$ .

However, due to the fact that  $\hat{b}$  contains noise Eq. 3.16 is incorrectly posed and  $A$  will be ill-conditioned. Hence, the elements of  $A^t$  are large and the error in  $\hat{b}$  will be magnified, obscuring the true solution which is

$$c = A^t b \quad (3.18)$$

Aulick and Gallie (1981) point out that to avoid this problem one needs to calculate solutions to Eq. 3.16 in a way which will damp out the error in  $\hat{b}$ . One way of accomplishing this goal is to supply some additional a priori information about the shape of the desired vector  $c$ . The additional information recasts the problem as a constrained optimization problem. This technique is called regularization.

An example of the type of a priori information commonly supplied appears in the deconvolution technique used by Reddy and Carrigan (1970). In their approach the resolution function is assumed to be the sum of two Gaussians with the same centroid but differing in their amplitudes and widths. The shape of  $f(x)$  (returning to the more common notation) is assumed to be a truncated parabola based on the free electron model (Reddy and Carrigan 1970).

Introducing these constraints on the shapes of  $r(x)$  and  $f(x)$  into the least square solution problem yields a physically plausible result. That is, the function  $f(x)$  which is obtained by this method does not contain indefinitely large and rapidly oscillating components typical of the results obtained by evaluating Eq. 3.8 directly. The difficulties associated with this method are related to the validity of the a priori assumptions. For example, the resolution function  $r(x)$  is constrained to be symmetric. In addition assuming the shape of the true Doppler energy distribution,  $f(x)$ , to be a simple truncated parabola is probably an oversimplification. Other investigators

(Mackenzie et al. 1976) suggest the modeling of  $f(x)$  using the superposition of Gaussian and inverted parabolic functions as representing contributions from core and valence electrons respectively. While it is not the purpose of this work to discuss the validity of the various electron distribution models, the very fact that there exists more than one analytic prediction of the shape of  $f(x)$  is strong reason to develop a deconvolution technique which places minimal constraints on the shape of  $f(x)$ .

One of the most widely used deconvolution techniques in Doppler PAS studies is an iteratively based method introduced by Jackman et al. (1974). In this method an initial estimate of the desired result  $f(x)$ , say  $f_1(x)$ , is selected. The estimate  $f_1(x)$  is then convoluted with  $r(x)$  to obtain  $h_1(x)$ , the first estimate of the experimentally obtained energy distribution  $h(x)$ . The function  $h_1(x)$  is then compared point-by-point with  $h(x)$  and if satisfactory agreement is obtained  $f_1(x)$  is assumed to be a reasonable approximation to the true solution. If unsatisfactory agreement is obtained then  $f_1(x)$  is modified to produce  $f_2(x)$  and the process is repeated.

The initial estimate of  $f(x)$ , the modification procedure for obtaining  $f_2(x)$  from  $f_1(x)$ , and the criteria for stopping the iteration procedure are all current topics of investigation in the literature. In the original method the experimentally obtained function  $h(x)$  served as the initial estimate  $f_1(x)$ . The modification algorithm was

$$f_2(x_i) = f_1(x_i) \times \left( \frac{\sum_{j=i-2}^{i+2} h(x_j)}{\sum_{j=i-2}^{i+2} h_1(x_j)} \right) \quad (3.19)$$

Subsequent work demonstrated that the convergence rate of the iteration process could be improved if the summations in Eq. 3.19 were taken over nine channels instead of the original five and if the summation ratio was squared before multiplication by  $f_1(x_j)$  (Dannefaer and Kerr 1975). The iteration pro-

cedure is stopped when the comparison of  $h(x)$  and  $h_N(x)$  yields a  $\chi^2$  value less than some predetermined minimum (Jackman et al. 1974) or when the sum of the squares of the deviations falls below some predetermined level (Dannefaer and Kerr 1975).

The main advantage of these iteratively based techniques is that a solution is obtained without any restriction on the shape of  $f(x)$ . While this is indeed an attractive feature of the method it also suffers from some inherent disadvantages. These disadvantages include the amount of computer time required, the possible dependence of the result on the initial estimate of  $f(x)$ , and the possibility of introducing large oscillations into the solution if the stopping criterion is selected with too small an error bound. This last difficulty is of course related to the mathematical nature of the incorrectly posed problem.

The iterative deconvolution technique essentially ignores the statistical nature of the problem. An alternate approach is to treat Eq. 3.4 as a problem of mathematical statistics and introduce a regularizer (a priori information) in the form of some probability distribution for  $f(x)$  (Turchin et al. 1971). This is the method of statistical regularization which offers the advantage of error analysis. The result of this process is the most probable  $f(x)$  for a given  $h(x)$  measured with mean square errors  $S(x)$  which are independent of  $x$  and normally distributed with zero mean (Goldanskii et al. 1978). The regularizer is a form of a priori information concerning the shape of  $f(x)$  and hence somewhat restrictive. However, such an approach appears to yield reasonable results if a solution can be obtained using minimal a priori information concerning the smoothness of  $f(x)$ .

In 1981 the Fourier cut-integral method was introduced by Szotek and Chmielewski. In this technique Eq. 3.4 is rewritten as:

$$f(x) = \frac{1}{2\pi} \int_{-\rho_0}^{\rho_0} e^{iwx} \frac{H(w)}{R(w)} dw \quad (3.20)$$

The difference in the equations is the introduction of finite limits of integration. If  $\rho_0$  is then chosen small enough the solutions obtained are physically plausible. The mathematical explanation for this result is that the insertion of finite limits of integration forces the dispersion of  $f(x)$  to remain finite. (Refer to Eq. 3.12 and the associated discussion). The problem associated with this method is that the optimum value of  $\rho_0$  is not readily predictable. Approximations for  $f(x)$  are obtained using different values for  $\rho_0$ , and one must then select the "best" approximation to the true  $f(x)$ . This is typically done by convoluting the function with  $r(x)$  and then comparing the result with  $h(x)$ . As  $\rho_0$  becomes too large the solution begins to exhibit large oscillations as would be expected from the mathematical theory.

In 1982 Verkerk published an extensive mathematical treatment of the deconvolution problem. In this work he discusses the relative strengths and weaknesses of many of the currently employed deconvolution techniques. Verkerk then proposes a method which in many ways is very similar to the Fourier cut-integral method. The crux of this method is that the high frequency Fourier coefficients of  $h(x)$  do not contain significant information and consequently only a limited number (say  $K$ ) of Fourier coefficients  $H(w)$  contain the essential information from measurement of  $h(x)$ . It should be pointed out that if  $H(w)$  contains  $N$  Fourier coefficients then deleting the  $N-K$  high frequency coefficients in the reconstruction of  $f(x)$  is equivalent to inserting finite limits of integration in Eq. 3.4 as in the Fourier cut-integral method.

The deletion of the high frequency coefficients is equivalent to a rectangular low-pass filter and it is well known that the application of such

a filter to  $F(w)$  will cause nonphysical oscillations in the approximation to  $f(x)$ .

In summary, there are two different types of errors associated with the estimate of  $f(x)$ : 1) statistical errors, which are amplified by the deconvolution operation and 2) systematic errors due to filtering, which are largest in the peak region and may cause nonphysical oscillations in the tails of the deconvoluted signal. Error type 1 increases as the number of Fourier coefficients used in reconstruction increases, while error type 2 decreases (Verkerk, 1982). Thus, by using such an approach the problem is reduced to selecting the optimum number of coefficients. One disadvantage of Verkerk's method, as with the Fourier cut-integral method, is that the procedure must be repeated several times for different numbers of coefficients until the optimum number is determined. It would be desirable to develop a simple one-step method to determine the appropriate number of coefficients to use in reconstructing  $f(x)$  from  $F(w)$ , and indeed this is one of the primary advantages of the FFT/power spectrum deconvolution method which is described in the following section.

#### The FFT/Power Spectrum Deconvolution Method

The deconvolution method developed in this work is a fast Fourier transform (FFT) based method which is simple to execute, fast, easily automated, and assumes little or no a priori information concerning the shape of  $f(x)$ . The distinctive feature of this method is that the number of Fourier coefficients retained in the reconstruction of  $f(x)$  is determined by examining the power spectrum of the experimental data. Using the observation that the power spectrum falls steeply to a minimum before rising due to the white noise component in the experimental data, the method truncates the Fourier

coefficients at this first minimum. The logic behind this approach is rather simple. Retaining fewer coefficients than this implies a smoothing of the deconvoluted spectrum and the loss of potentially valuable structural information. Adding more coefficients gains nothing because the signal-to-noise ratio is of order one at the minimum. In addition, truncating the coefficients at the point where the white noise begins to dominate forces the dispersion of  $f(x)$  to remain finite.

With this brief introduction it is now possible to list the six steps which together constitute the FFT/power spectrum deconvolution method:

- 1) Both the raw data  $h(x)$  and the resolution function  $r(x)$  are background corrected.
- 2) The functions  $h(x)$  and  $r(x)$  are Fourier transformed using an appropriate FFT algorithm.
- 3) The Fourier coefficients of  $h(x)$  are examined and those up to and including the first minimum in the power spectrum of  $h$  are retained, the rest are replaced by zeros.
- 4) The Fourier transform of  $f(x)$  is obtained by the point by point division of the Fourier transform of  $h(x)$  by the transform of  $r(x)$ .
- 5)  $f(x)$  is obtained by the inverse FFT.
- 6) The solution is checked by forming the convolution of  $f(x)$  with  $r(x)$  and comparing the result with  $h(x)$ .

Each of these basic steps will now be described in further detail.

Semiconductor detectors usually produce asymmetric  $\gamma$ -ray peaks with a non-linear underlying background. This is caused primarily by three processes:

- 1) incomplete charge collection, 2) electron escape from the active volume, and 3) scattering of  $\gamma$ -rays (Chaglar et al. 1981). This background component

~~should be removed from the experimentally obtained energy distributions before~~

the deconvolution procedure is performed. The background correction routine employed in this work involves a least squares fitting of a linear function in both the low and high energy tail regions. The region on the low energy side of the peak spans from 486 to 497 KeV, while the high energy region extends from 534 to 542 KeV. The straight line segments fitted in these regions are extrapolated to 503 and 518 KeV respectively where they are joined by a third line segment to produce a simple background approximation (Jackman et al. 1974). The linear least squares analysis algorithm used in this work is a simplification of a higher order least squares routine by Gerald (1978). As in the work of Jackman et al. (1974), use is made of only the high energy half of the background corrected spectrum since it is known that incomplete charge collection of solid state detectors results in low energy tailing. In theory the spectrum should be symmetric about 511 KeV since statistically one should observe equal numbers of red and blue Doppler shifted  $\gamma$ -rays.

Before proceeding to the details of the FFT algorithm one important point requires explanation. In order to correlate MCA channels with a corresponding energy range, one must know the MCA energy resolution per channel as well as which channel corresponds to the center of the energy distribution and hence 511 KeV. The former value is determined as described in the experimental procedures. However, the center of the distribution must be calculated for each experimental spectrum. A simple centroid calculation is unacceptable since the higher background on the low energy side would bias the result. Similarly, simply selecting the channel with the maximum number of counts is undesirable since, due to the statistical nature of the experiment, the centroid channel and the peak channel are not necessarily the same. The technique used in this work is to determine the eleven channel region which contains the largest number of counts and call the middle channel of this region

the center of the distribution. In most cases, the channel corresponding to the center as calculated above was also the channel with the maximum number of counts.

Proper arrangement of the data string is required before proceeding to the FFT algorithm. Since this algorithm requires periodic data, the raw data are rearranged in the following way. The first datum point of the rearranged set corresponds to the center of the raw data string. The remaining rearranged data points numbered from 2 to  $((N/2) + 1)$  correspond to the remaining values of the background corrected high energy half of the raw spectrum. It should be pointed out that  $N$  must satisfy the relationship  $N = 2^m$  where  $m$  is an integer. Since the real data string rarely has exactly  $((N/2) + 1)$  data values it is usually necessary to insert a string of zeros at the end of the data set to fulfill this requirement. In order to achieve periodicity, data point  $N$  should correspond to data point 2. In general the points from  $((N/2) + 2)$  to  $N$  are defined in the following way:

$$g((N/2) + P) = g((N/2) + 2 - P) \quad (3.21)$$

where  $P$  varies from 2 to  $(N/2)$ . In the experiment performed in this work  $m$  was equal to eleven which corresponds to a total of 2048 data points (including the zerofill).

Once the functions  $h(x)$  and  $r(x)$  have been background corrected and properly arranged they are passed to the FFT algorithms. The FFT algorithms employed in this work are those developed by Cooley, Lewis, and Welch (1979). There are a number of other such algorithms available which should work equally well. In every case it is recommended that the Fourier coefficients be calculated with double precision accuracy. The advantage of using FFT's in place of discrete Fourier transforms (DFT) can be seen from the following example. For 2048 data points the DFT requires  $N^2$  or  $\sim 4.2 \times 10^6$  complex mul-



tiplifications while the FFT requires  $2N \log_2 N$  or  $\sim 4.5 \times 10^4$  complex multiplications. In this example the number of calculations required is reduced by about two orders of magnitude.

The justification for using only the Fourier coefficients up to and including the one corresponding to the first minimum in the power spectrum of  $h(x)$  has been briefly discussed. Similar techniques are commonly employed in electronic signal processing and it can be shown that the number of coefficients retained is a measure of the frequency resolution. In the present case the resolution refers to the width of the lineshape components which can be resolved. The procedure by which the computer calculates the number of retained Fourier coefficients from  $H(w)$  is as follows. Values associated with the power spectrum are obtained by multiplying each Fourier coefficient  $H(K)$  by its complex conjugate. However, since the function  $h(x)$  is even, the Fourier coefficients  $H(w)$  are all real. Truncating  $H(w)$  at the  $K$  corresponding to the first minimum in the power spectrum is equivalent to the requirement that the set of retained coefficients be monotonically decreasing, which is easily programmed by testing

$$H(w + 1) < H(w) \quad (3.22)$$

In this analysis only the first  $(N/2) + 1$  Fourier coefficients are considered. The first coefficient corresponds to the D.C. component of zeroth frequency and the  $N/2 + 1$  coefficient corresponds to the highest frequency available. It is well known that the remaining coefficients correspond to the negative frequencies and are in fact the complex conjugates of the first  $(N/2 + 1)$  coefficients following the relationship  $g(N/2 + P) = g^*(N/2 + 2 - P)$  for  $P = 2$  to  $N/2$ . However since all the coefficients are real, no new information is to be gained by examining the negative frequencies.

Once the appropriate number of coefficients, say  $I$ , has been determined the remaining  $N-I$  coefficients of  $H(w)$  are set equal to zero before dividing  $H(w)$  point-by-point by  $R(w)$  and proceeding to obtain  $f(x)$  by the inverse Fourier transform. The consequences of this truncation, or filtering operation are well known. See, for example, the discussion by Verkerk (1982). In general the process results in some systematic errors in which the peak values are underestimated and nonphysical oscillations may appear in the deconvoluted function  $f(x)$ . For Doppler-broadened positron annihilation spectra taken with our instrumentation this filtering error is negligible for Doppler spectra having more than 40K peak counts.

To check the accuracy of the function  $f(x)$  requires the convolution of this function with  $r(x)$  and the comparison of the resulting function with  $h(x)$ . The details of practical convolution techniques have been discussed by Lam *et al.* (1981), and the procedure used in this work is essentially the same.

A complete listing of the Fortran FFT/power spectrum deconvolution program is given in Appendix F.

### Test of the Method

In order to explore the validity of the FFT/power spectrum method the following test was performed. The resolution function  $r(x)$  was obtained from the 514 KeV gamma ray of  $^{85}\text{Sr}$ , while the function  $h(x)$  was measured for a neutron irradiated high purity single crystal sapphire sample. This sample had been irradiated in EBR-II at 740°C to a fluence of  $3 \times 10^{21}$  neutrons/cm<sup>2</sup> with  $E < 0.1$  MeV. Both  $r(x)$  and  $h(x)$  were then processed as described in the previous section. For this test of the method  $f(x)$  was estimated from  $F(w)$  using the first 101 low frequency Fourier coefficients, as selected by power spec-

tral analysis.

Figure 5 is a plot of the power spectrum for both  $F(w)$  and  $H(w)$ . The filled circles represent the spectrum of the experimentally obtained function  $h(x)$ . Note the rapid decrease of about six orders of magnitude in power with increasing frequency. This power spectrum is about what would be expected from a wide-band-width stochastic process contaminated with white noise. This white noise component is associated with the band of frequencies to the right of the first minimum in the power spectrum of  $h(x)$ . Although the overall signal-to-noise ratio is very high, for frequencies in this white noise band the signal-to-noise ratio is less than one and therefore it is impossible to retrieve any structural information about the function  $f(x)$  in this high frequency range. This suggests that only those coefficients to the left of the white noise band, that is to the left of the first minimum, be retained. To use less than this number of coefficients in reconstruction is undesirable since it is known that the errors associated with truncation, or filtering, increase with a decrease in the number of coefficients used.

The amplification of the statistical errors in  $h(x)$  by the deconvolution procedure can be clearly seen in Figure 5. The open circles represent the spectrum of the deconvoluted function  $f(x)$ . Note the drastic increase in the power associated with the high frequencies of  $f(x)$  as compared with the corresponding frequencies in  $h(x)$ . However, the low frequency coefficients to the left of the first minimum are well behaved. These observations are the foundation on which the FFT/power spectrum method is based.

It is not surprising to find that the number of retained Fourier reconstruction coefficients, as determined by power spectrum analysis, is a function of the number of peak counts in the raw data  $h(x)$ . Figure 6 is a semi-log plot of this relationship for irradiated sapphire. This figure sug-

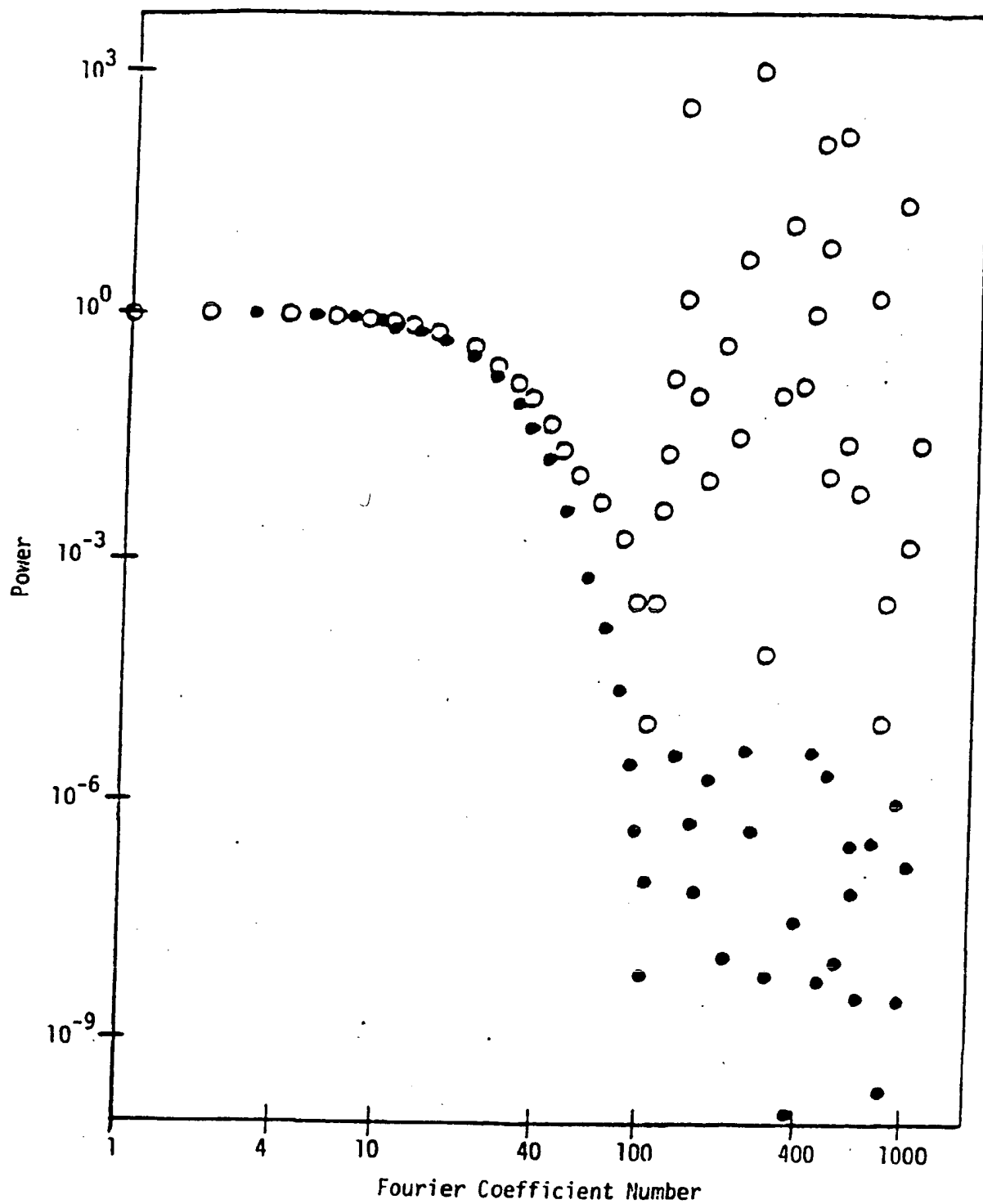


Figure 5. A Plot of the Power Spectrum of the Raw Data,  $h(x)$ , (Filled Circles), vs. the Deconvoluted Data,  $f(x)$ , (Open Circles). Note that increasing Fourier coefficient number corresponds to increasing frequency.

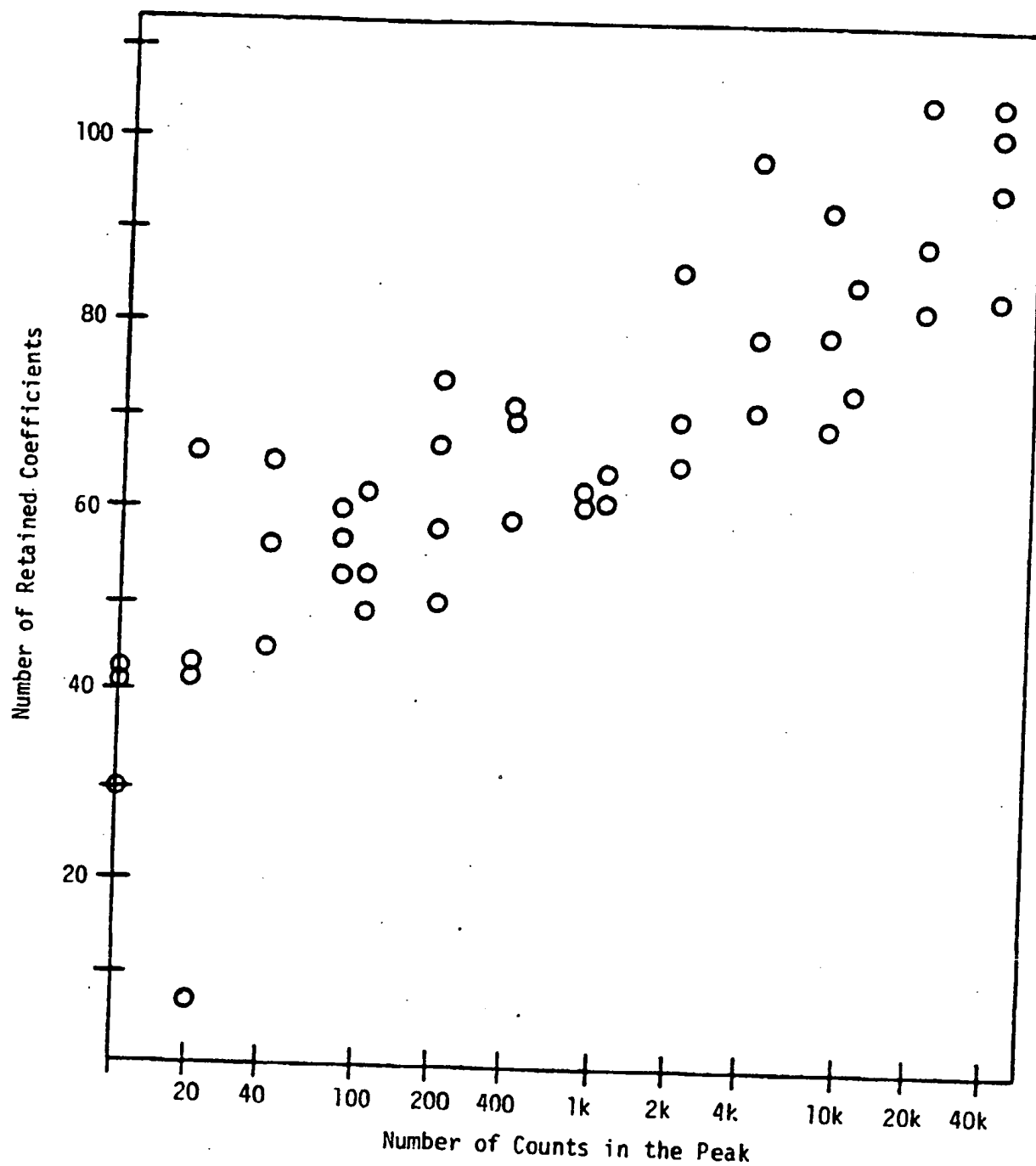


Figure 6. A Semi-Log Plot of the Number of Reconstruction Coefficients Retained by the FFT/PS Deconvolution Algorithm as a Function of the Number of Counts in the Peak Channel of the Raw Data. The Data is for Irradiated Sapphire.

gests that the relationship over this range of peak counts is an exponential of the form:

$$y \propto m \ln x \quad (3.23)$$

where  $y$  is the number of coefficients,  $x$  is the number of counts in the peak, and  $m$  is approximately equal to 7 for this data set. This curve demonstrates the high price one must pay in terms of data collection time to increase significantly the number of retained coefficients. Fortunately, our experience has shown that with the ~100 coefficients which are usually obtainable with 40K counts in the peak (30 minutes of data collection) the results obtained are very satisfactory.

Figure 7 is a comparison of the deconvoluted function  $f(x)$  and the raw data  $h(x)$  after background correction. The curves have been normalized to equal area. As expected, the deconvoluted curve is both sharper and smoother than the raw data. There appears to be a slight oscillation in  $f(x)$  in the tail region. The quality of this estimate of  $f(x)$  can be examined in the usual way by convolution with  $r(x)$  to obtain an approximation  $h'(x)$  to the original experimental function  $h(x)$  as is shown in Figure 8. The overall quality of the fit is so good that on this scale it is difficult to see any differences between the raw spectrum and the convoluted spectrum. The one exception occurs in the peak region where several of the raw data points of  $h(x)$  lie well below their counterparts in  $h'(x)$ . Careful examination of the shape of  $h(x)$  in this region reveals some rather spurious changes in the slope of the spectrum. Physical constraints on the annihilation process suggest that, assuming no positronium formation, the annihilation spectrum should be relatively smooth in this region. This suggests that these slope changes in  $h(x)$  are related to statistical counting errors. To test this idea two other

One of the theoretical advantages of deconvolution is a significant improvement in the resolution of DBPAS measurements. Figure 9 is a plot of the high energy half of a typical  $^{85}\text{Sr}$  peak before and after deconvolution. The closed circles represent the raw data after background correction, while the open circles show the shape of the resolution curve after deconvolution using 243 coefficients. This is the number of coefficients selected by the FFT/PS method corresponding to the first minimum in the power spectrum for this data set. Note that the full width half maximum (FWHM) value for the raw data is  $\sim 1280$  eV while the deconvoluted curve has a FWHM of  $\sim 320$  eV. This corresponds to a factor of four improvement in resolution. However, this result is somewhat misleading in that for aluminum the number of retained coefficients is approximately 60-70. The triangles in Figure 9 show the resolution function after deconvolution using 70 retained coefficients. In this case the FWHM is  $\sim 576$  eV corresponding to an improvement in resolution by a factor of  $\sim 2.2$ . Clearly it can be seen that the improvement in resolution obtained by deconvolution using the FFT/PS method is a function of the number of retained coefficients, which in turn is directly related to the statistical quality of the raw data set (Schaffer et al. 1984). This implies that the data collection process should be carried out for sufficiently long times in order to obtain good counting statistics. (The counting error is proportional to  $1/\sqrt{N}$ , where  $N$  is the number of counts in the channel).

Unfortunately, counting for long times introduces another error source. This error source is related to instrumental electronic drift due, at least in part, to temperature fluctuations. There are two major ways in which other researchers have dealt with this problem. First, the accuracy of results can be improved by operating the entire DBPAS system within a temperature controlled ( $\pm 0.2^\circ\text{C}$ ) atmosphere. The second method, usually used in

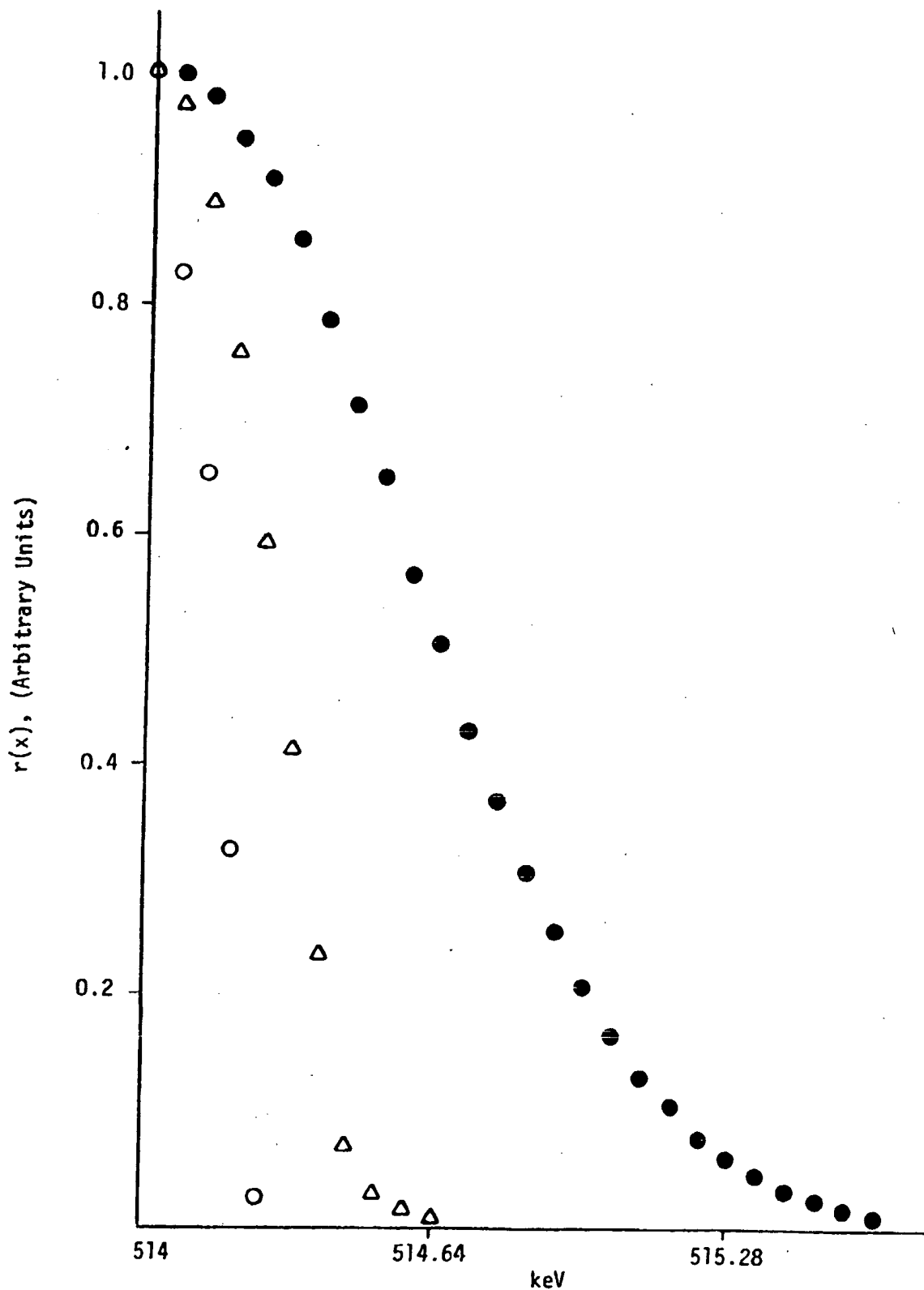


Figure 9. A Comparison of the Shape of the High Energy Half of the Resolution Function,  $r(x)$ , a) Before Deconvolution, ( $\bullet$ ), b) After Deconvolution using 70 Coefficients ( $\Delta$ ), and c) After Deconvolution using 243 Coefficients, ( $\circ$ ).



conjunction with the first, is to digitally stabilize the electronics. This digital stabilization is accomplished by simultaneously monitoring the 511 KeV annihilation line along with a suitably located monoenergetic gamma ray source such as the 478 KeV gamma ray from  $^7\text{Be}$ . In this procedure a spectrum is collected for  $\sim 100\text{S}$  and then the centroid of the 478 KeV peak is calculated. Subsequently, several other spectra are collected and their corresponding centroids also calculated. Next, each spectrum is linearly shifted such that all of the 478 KeV centroids coincide. Finally the 511 KeV peaks are summed to produce a DBPAS spectrum with as many total integrated counts as is necessary to obtain suitable counting statistics.

While temperature and/or digital stabilization should improve the reproducibility of DBPAS measurements, it is still possible to obtain meaningful results using unstabilized equipment. In this work neither of the stabilization techniques were employed, and the major consequence of such a procedure is an observable spread in the values of the lineshape parameters calculated for repeated DBPAS measurements of the same metallurgical sample. This problem will be discussed in greater detail in the sections of this chapter dealing with the separation of multiple defect types.

In order to minimize the change in the metallurgical defect structure of the samples as a function of time, all of the DBPAS measurements in this work were made at  $77^\circ\text{K}$ . Performing the measurements at liquid nitrogen temperatures raises an important theoretical question: is the assumption that the positrons are essentially at rest, with respect to the electrons, at the time of annihilation still valid? Previous experiments by Kubica and Stewart (1975) demonstrate that even at temperatures as low as  $4.2^\circ\text{K}$  the positrons reach thermal velocities prior to annihilation.

In order to determine the effect of measuring the annihilation

lineshape at 77°K an experiment was performed in which six DBPAS spectra were obtained for a well annealed pair of aluminum samples. Three of the measurements were made at 77°K and three at room temperature. The small deviation (of order  $1/\sqrt{N}$ ) between the six spectra showed no correlation with measurement temperature. It was therefore concluded that decreasing the sample temperature from room temperature to 77°K would not affect the positron-electron annihilation procedure in any experimentally observable manner.

As mentioned in the experimental procedures the samples examined in this work were high purity aluminum disks heat treated to control defect type and concentration. It is assumed in the remainder of this work, without direct TEM evidence but based on the arguments presented in the experimental section, that one group of samples contained predominately monovacancies while a second group of samples possessed a defect structure dominated by prismatic dislocation loops. For each pair of disks three or four annihilation energy spectra were obtained, from which an array of lineshape parameters were subsequently extracted numerically. The lineshape parameters examined in this work include the first five even central moments of the energy distributions defined by the relationship:

$$m(I) = \frac{1}{N-1} \sum_{i=1}^N (x_i - \bar{x})^I f(x_i) \quad (4.1)$$

where  $N$  is the number of channels in the spectrum

$\bar{x}$  is the channel corresponding to the spectrum centroid

$f(x_i)$  is the number of counts in channel  $i$

and  $I$  is an even integer (2,4,6,8,10) corresponding to the first through the fifth even central moments

A second group of parameters is defined by the relationship:

$$x(I) = \left[ \sum_{i=j}^{j+4} f(x_i) \right] / \sum_{i=\bar{x}}^{\bar{x}+59} f(x_i) \quad (4.2)$$

with  $j$  defined as  $j = \bar{x} + 5(I-1)$

where  $f(x_i)$  is the number of counts in channel  $i$

$\bar{x}$  is the channel corresponding to the spectrum centroid

and  $I$  is an integer from 1 through 12

For example,  $x(1)$  is the ratio of the number of counts in the first five channel regions on the high energy side of the centroid (actually it includes the centroid) to the total number of counts in the high energy half of the spectrum. Similarly,  $x(12)$  is the ratio of the number of counts in the twelfth five channel interval (to the right of the centroid) to the total number of counts in the high energy half of the spectrum.

The third, and final, group of lineshape parameters examined are the conventional "S" parameters (MacKenzie et al. 1970). These parameters are defined as the ratio of the number of counts in some central energy region to the total number of counts in the energy spectrum. The width of the energy window used to define the S parameter is selected so that the value of S is in the range 0.4 - 0.6 (Smedskjaer and Fluss 1983). In this study three S-parameters (S(1344), S(1472), and S(1600)) were examined with respective central energy windows of 1.344, 1.472, and 1.600 KeV.

These parameters were calculated for both the background corrected raw data and for the deconvoluted data. Hence a total of forty lineshape parameters were calculated for each DBPAS measurement.

Figure 10 is a plot of the parameter  $x(1)$  for the deconvoluted data. The open circles represent the values of this parameter for the "as quenched" samples (predominately monovacancies). The filled circles correspond to the

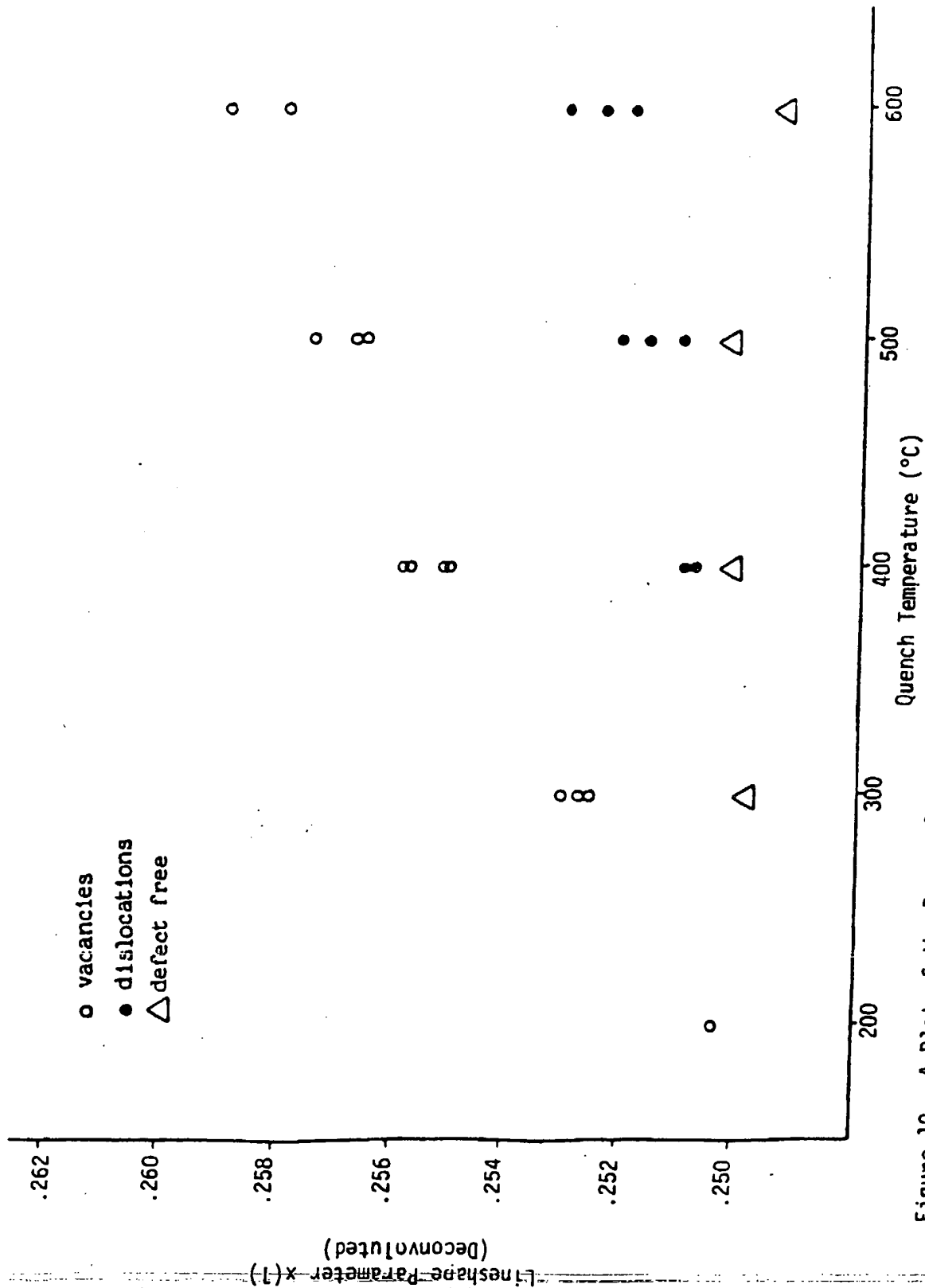


Figure 10. A Plot of the Deconvoluted Lineshape Parameter  $x(1)$  as a Function of Quench Temperature.

low temperature anneal samples (prismatic dislocation loops), and the triangles represent the samples after the high temperature anneal (the essentially defect free state). Note that the numerical value of  $x(1)$  increases with quenching temperature for both defect types. This trend is in agreement with theoretical predictions since an increasing defect concentration results in a higher probability of annihilation with lower kinetic energy conduction electrons, corresponding to a more narrow energy spectrum, and hence a larger value for  $x(1)$ . The equations used to predict the theoretical defect concentrations for both the vacancies and the prismatic dislocation loops as a function of quenching temperature are discussed in Appendix G.

Figure 10 demonstrates that DBPAS measurements are sensitive to the presence of both vacancies and dislocations in high purity aluminum. This result is, of course, a necessary prerequisite for the possibility of DBPAS measurements being able to distinguish between the defect types. From this plot it is possible to perform some semiquantitative analysis and estimate some physical properties of aluminum. For example, one can estimate values of the "cross section" for the trapping of a positron by both vacancies and dislocations in aluminum. The accuracy of these estimates is limited by the reproducibility of the DBPAS measurements as well as the need for some independent information such as the lifetime of positrons as a function of defect type.

In 1972 Cotterill et al. published a paper in which they examined the average positron lifetime,  $\tau_{avg}$ , on a set of aluminum samples very similar to the ones studied in this work. Their work clearly demonstrated that positron lifetime experiments were sensitive to both vacancies and dislocations in aluminum. Before discussing the similarities between their work and the ~~current experiments it is important to demonstrate the relationship between~~

the parameters  $\tau_{avg}$  and  $x(1)$ . In a review article by Sieger (1980) it is shown that:

$$\tau_{avg} = (1 - A_t)\tau_b + A_t\tau_t \quad (4.3)$$

where  $\tau_b$  is the lifetime in the bulk material

$\tau_t$  is the lifetime in the trapped state

and  $A_t$  is the probability that the positron annihilates from the trapped state

Similarly, Sieger demonstrates that:

$$F = (1 - A_t)F_b + A_tF_t \quad (4.4)$$

where  $F$  is any characteristic shape parameter obtained from the analysis of DBPAS measurements

$F_b$  is the value of the parameter in the bulk state

and  $F_t$  is the value of the parameter in the trapped state

Since the parameter  $x(1)$  can be shown to be equivalent to an "S" type parameter, and since an "S" type parameter is a common characteristic line-shape parameter, Eq. (4.4) can be rewritten as:

$$x(1) = (1 - A_t)x(1)_b + A_tx(1)_t \quad (4.5)$$

where  $x(1)_b$  is the value of the parameter in the bulk state

and  $x(1)_t$  is the value of the parameter in the trapped state

Clearly a comparison of Eq. (4.3) and Eq. (4.5) demonstrates the similarity between  $\tau_{avg}$  and  $x(1)$ .

With this in mind it is not surprising to find that plots of  $\tau_{avg}$  vs. quench temperature ( $T_Q$ ) and  $x(1)$  vs.  $T_Q$  share many common characteristics. (The  $\tau_{avg}$  plot used for comparison is Figure 2 in the work of Cotterill et al., 1972). In both figures the  $T_Q$  at which the vacancy concentration is large enough to affect the measured parameter is  $\sim 200^\circ\text{C}$ . The saturation value of both  $\tau_{avg}$  and  $x(1)$  for vacancies occurs at  $T_Q \sim 600^\circ\text{C}$ . The curve through

the points corresponding to vacancies is sigmoidal in shape for both the lifetime and deconvoluted DBPAS measurements. As for the points corresponding to samples with prismatic dislocation loops, the value of both parameters initially deviates from their bulk values for samples annealed after a quench from  $\sim 400^\circ\text{C}$ . Neither parameter reaches a saturation value for dislocations within the temperature range examined in this work.

Figure 10, therefore, shows not only the ability of deconvoluted DBPAS measurements to detect the trapping of positrons by both vacancies and dislocations in aluminum, but also qualitatively verifies the theoretically predicted equivalence of the lifetime parameter  $\tau_{\text{avg}}$  and the Doppler parameter  $x(1)$ .

Figure 11 is a plot of the same lineshape parameter,  $x(1)$ , vs.  $T_Q$  for the background corrected raw data, i.e. the same data as shown in Fig. 10 but before the deconvolution procedure. Note the general similarity between these two figures. Careful examination of the raw data shows that the points corresponding to the  $400^\circ\text{C}$  quench are systematically displaced. That is, the entire group of five data points seems to be "too high" as compared with the rest of the data. To understand the cause of this shift it is necessary to examine the shape of the instrument resolution function  $R(x)$ .

During the course of these experiments the instrument resolution was periodically checked by collecting an energy spectrum for  $^{85}\text{Sr}$ . The monoenergetic line is broadened due to the physical characteristics of the DBPAS system as previously described. This broadened energy line serves as the resolution function,  $R(x)$ , in the FFT/PS deconvolution procedure. It also describes the amount by which the undistorted energy spectrum is broadened by the instrumentation.

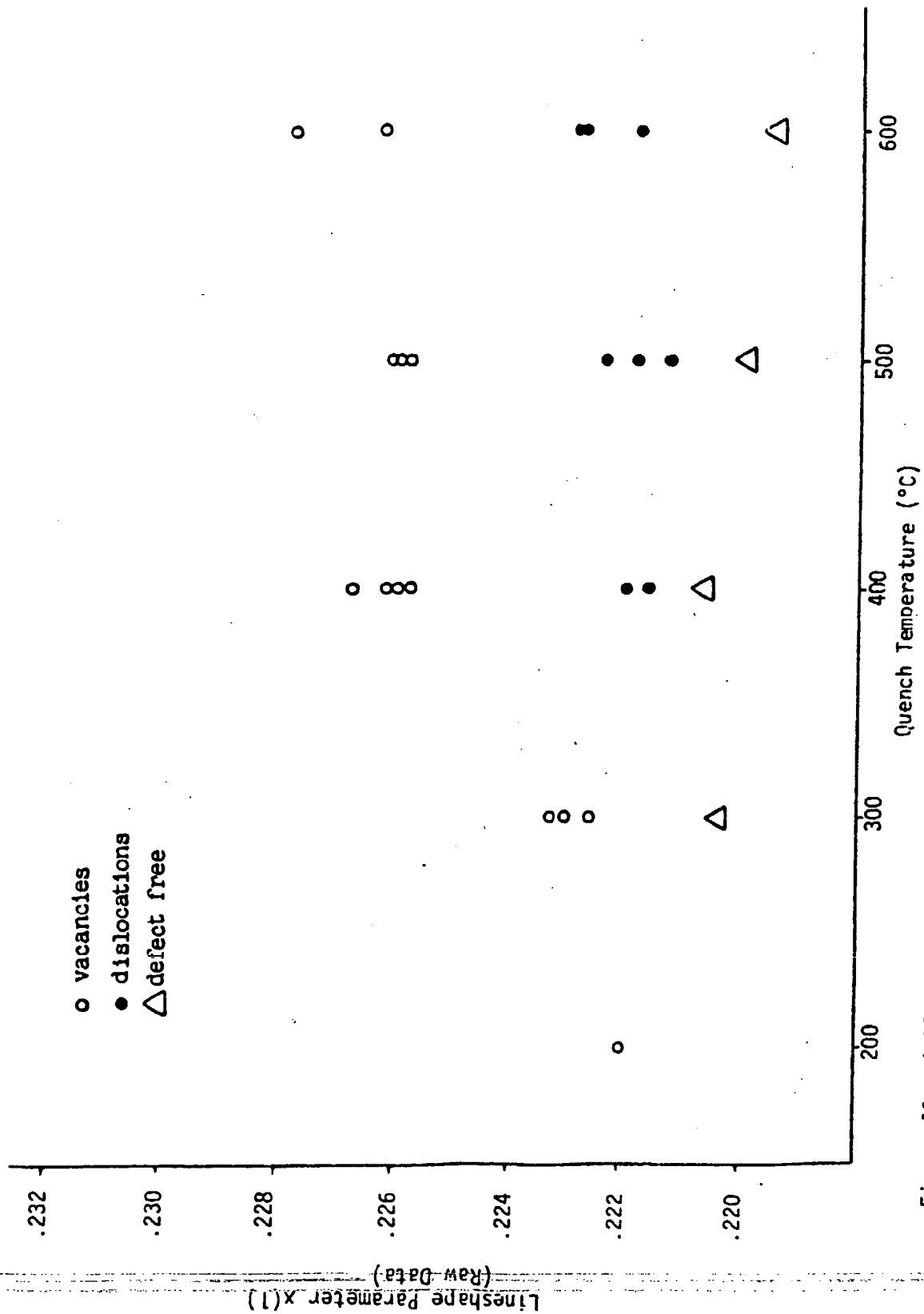


Figure 11. A Plot of the Raw Data Lineshape Parameter  $x(1)$  as a Function of Quench Temperature.



Figure 12 shows two such resolution spectra. The open circles represent the instrument resolution corresponding to the DBPAS measurements for the samples quenched from 400°C. The filled circles represent a resolution function which is characteristic (within statistical error) of the resolution functions for all of the remaining data points. This comparatively large shape change in the function  $R(x)$  was apparently caused by a power failure in the laboratory. Note that the Function  $R(x)$  corresponding to the 400°C samples (henceforth termed SR85OLD) is more sharply peaked, and therefore more narrow, than the other resolution function (SR85NEW).

The physical significance of the change in resolution function is as follows. An observed energy spectrum will be broadened in two ways. The first way is by a decrease in the concentration of vacancy type defects, in the sample being examined. The second broadening mechanism is, of course, the broadening due to the instrumentation. Now, if the second source of broadening remains essentially constant, then changes in the width of the observed spectra may be attributed to changes in defect concentration. On the other hand, if the broadening due to the instrumentation decreases, and this error source is not properly removed, the observed spectrum will be artificially narrow and its apparent defect concentration will be too great. Thus, if the data for the 400°C samples are collected with an artificially narrow resolution function, i.e. SR85OLD, their apparent defect concentration will be greater than they should be.

Of particular significance is, that while a change in experimental conditions can alter the shape of the resolution function and thereby distort the values of lineshape parameters calculated for the raw data, the deconvolution routine, when supplied with the appropriate  $R(x)$  function, automatically yields the desired results. This illustrates that for reproduc-

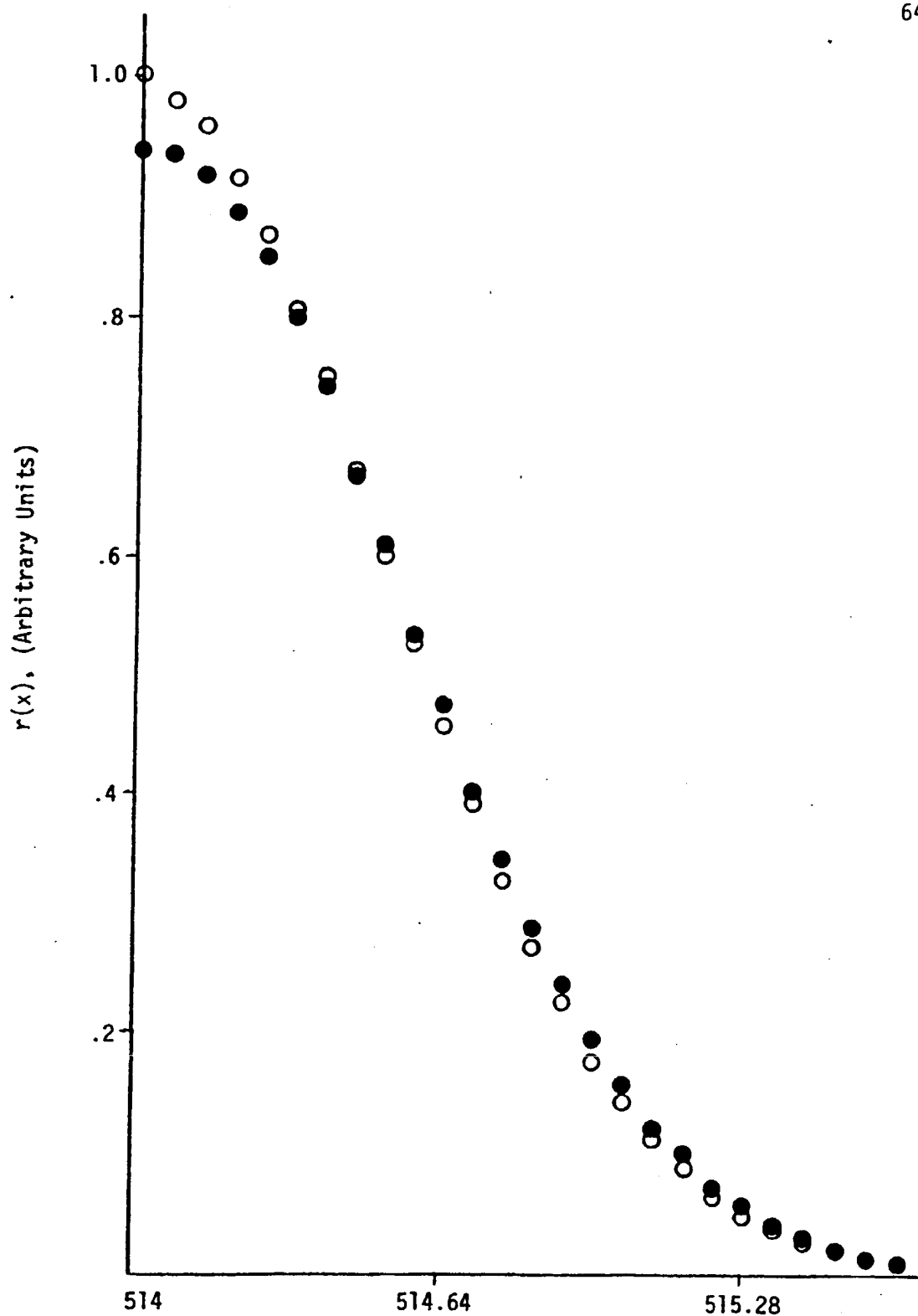


Figure 12. A Comparison of the High Energy Half of Two Resolution Functions,  $r(x)$ , Collected at Different Times During a Series of Experiments. The Two Functions are Referred to in the Text as SR35NEW ( $\bullet$ ) and SR85OLD ( $\circ$ ).

bility the deconvolution of DBPAS measurements should be considered. In addition, as mentioned previously, since deconvolution removes the instrumental effects it should yield lineshape parameters which are directly comparable between different laboratories.

Despite the advantages of deconvolution it is important to remember that this technique does, of course, have its limitations. In DBPAS measurements there are several sources of experimental error, each of which has a corresponding time scale. The magnitude of this time scale is the overriding factor in determining the applicability of deconvolution techniques in minimizing the specific error in question. For example, any change in experimental conditions which can be treated as a step function, that is, by changing from one constant value to another constant value in a very short period of time, can be easily treated by deconvolution. Such "error" sources include the accidental (e.g. power failure) or intentional changes in the electronic amplification of the energy signal. To correct for such a change one simply needs to collect a resolution function before and after the event and use the appropriate  $R(x)$  in the deconvolution algorithm.

On the other hand, error sources having a time scale approximately equal to the time required to collect a data set (tens of minutes) can not be corrected by deconvolution alone. Examples of such possible error sources include line voltage fluctuations, temperature variations, and electronic drift. These error sources may cause the centroid of the resolution function to vary during the collection period, resulting in a composite  $R(x)$  which is artificially broad. In addition, even if the raw data set to be deconvoluted and the function  $R(x)$  are collected in rapid succession the amount of centroid drift for the two energy spectra may well be uncorrelated. This type of error source requires a different means of correction. The DBPAS measurements

should be performed in a temperature controlled laboratory. Also, digital stabilization, as described previously, should be used to control centroid drift.

The final error type is the small time scale or high frequency class. An error source such as sixty-cycle electrical noise, if it were present, should average to zero over the time required to collect a data set. In addition, a high frequency error source should have a similar effect on both the resolution function and the DBPAS energy distribution.

In order to judge the effectiveness of deconvolution in eliminating experimental error it is necessary to define some quantitative measure of this error. The predominant manifestation of experimental error appears as a spread in lineshape parameter values calculated for multiple DBPAS measurements on the same samples. Figure 13 is a plot of the parameter  $x(1)$  vs.  $T_0$  for the deconvoluted data, note the spread in values of  $x(1)$  for a given defect configuration.

In an attempt to evaluate the performance of the deconvolution algorithm the following parameters are introduced (see Figure 13 for a physical interpretation).

- 1)  $\Delta P_V/P_0$  is a measure of the sensitivity of this parameter to the existence of vacancies.
- 2)  $\delta P_V/\Delta P_V$  is a measure of the error associated with the spread in values of this parameter for vacancies.
- 3)  $\Delta P_I/P_0$  is a measure of the sensitivity of this parameter to the existence of dislocations.
- 4)  $\delta P_I/\Delta P_I$  is a measure of the error associated with the spread in values of this parameter for dislocations.



Clearly, it is desirable for the sensitivity ( $\Delta P/P_0$ ) to be as large as possible and the error ( $\delta P/\Delta P$ ) as small as possible for both defect types. A comparison of these quantities calculated from the deconvoluted and the raw data is shown in Table 1. Note that for all four of the lineshape parameters listed the sensitivity ratio is higher and the error ratio is lower for the deconvoluted data than for the raw data. This analysis encourages the further use of the deconvolution procedure to improve the quality of the results obtained from DBPAS measurements.

A tentative explanation for the apparent improvement in sensitivity and accuracy of the deconvoluted results is based upon trends in the shape of the energy spectrum before and after the application of the FFT/PS analysis. Figure 14 shows the high energy half of the peak region for an Al sample quenched from  $\sim 600^\circ\text{C}$ . The deconvoluted spectrum is, of course, more narrow than the raw data but note that it is also smoother (due to the filtering of the high frequency components). One explanation for the comparatively large error ratios of the raw data is that they are related to the statistical counting errors in the peak region of the spectrum. If this is indeed the case it is not surprising to observe a decrease in the error ratio after the filtering of the high frequency components. However, the most probable explanation for the increased sensitivity resulting from deconvolution is related to the removal of the "broadening" action of the experimental instrumentation. Recall that the major effect of the instrumentation is to spread counts from one channel into several adjacent channels. Therefore, the true lineshape changes in the annihilation energy distribution, caused by the trapping of positrons at defects, are spread out over a much larger number of channels in the raw data. The deconvolution procedure corrects for this effect and in so doing restores some of the details of the "true" annihilation

Table 1

		<u>x(1)</u>	<u>x(2)</u>	<u>S(1344)</u>	<u>S(1600)</u>
DEC <sub>v</sub>	$\frac{\delta P_{\max}}{\Delta P}$	.111	.167	.183	.215
DEC <sub>v</sub>	$\frac{\Delta P}{P_0}$	.0360	.0263	.0325	.0279
DEC <sub>l</sub>	$\frac{\delta P_{\max}}{\Delta P}$	.315	.320	.331	.581
DEC <sub>l</sub>	$\frac{\Delta P}{P_0}$	.0124	.0110	.0119	.0106
ND <sub>v</sub>	$\frac{\delta P_{\max}}{\Delta P}$	.181	.235	.235	.231
ND <sub>v</sub>	$\frac{\Delta P}{P_0}$	.0348	.0165	.0256	.0207
ND <sub>l</sub>	$\frac{\delta P_{\max}}{\Delta P}$	.471	.938	.555	.585
ND <sub>l</sub>	$\frac{\Delta P}{P_0}$	.0155	.0077	.0100	.0078

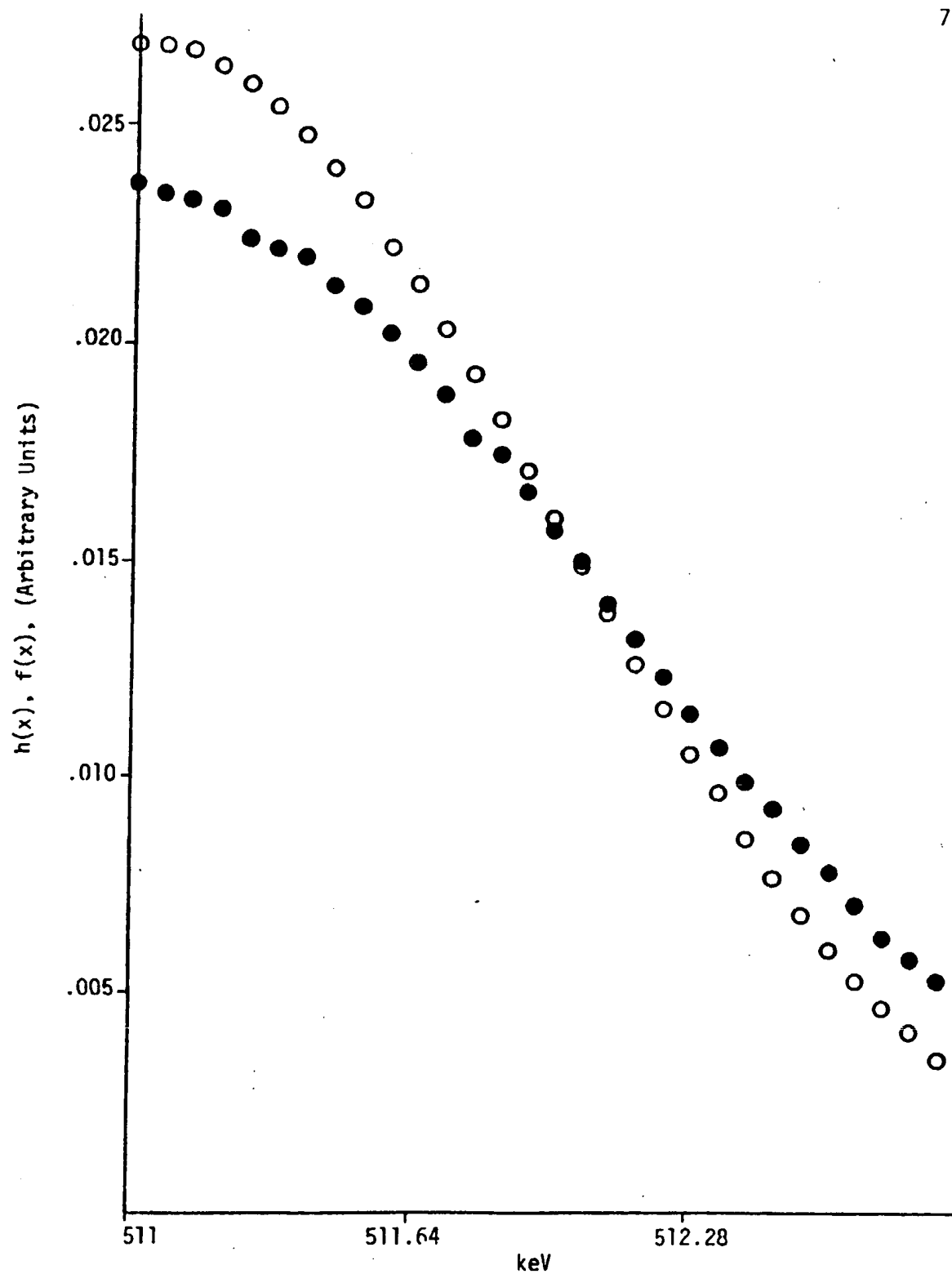


Figure 14. A Comparison of the High Energy Half of the Peak Region for  $h(x)$ , the Raw Data, (●), and  $f(x)$ , the Deconvoluted Data, (○), for an Aluminum Sample Quenched from 600°C.



energy distribution. The net result is an increase in defect sensitivity. It should be mentioned that the magnitudes of the enhancement in sensitivity and accuracy due to deconvolution noted in this work are consistent, but small. However, from this work alone it is not possible to estimate the effect of temperature and electronic drift upon the results. It would be extremely enlightening to repeat this analysis employing a temperature controlled and digitally stabilized system.

Before moving on from the topic of parameter sensitivity and accuracy, one more pertinent observation should be discussed. Figure 15 is a plot of the error ratio,  $\delta P/\Delta P$ , calculated separately at each quenching temperature (the parameter being examined is  $x(1)$ ). Several trends are apparent. First, it is clear that the error associated with the dislocations is larger than the error associated with vacancies. This trend may be a direct consequence of the size of the denominator,  $\Delta P$ , which is smaller, at a given quench temperature, for samples with dislocations than for the corresponding samples with vacancy type defects. This observation is however somewhat deceptive in that the horizontal axis of Figure 15 is  $T_Q$  and not defect concentration. A meaningful comparison of parameter sensitivity between defect types should be made using equivalent defect densities. Unfortunately, the dislocation densities which can be introduced by this thermal treatment are not large enough for direct comparison. In addition, it should be mentioned that the trapping of positrons by dislocations may be somewhat more complicated than trapping by vacancies. This idea is discussed in detail by Martin and Paetsch (1972). Very briefly, it may be that the depth of the potential well associated with a prismatic dislocation loop is such that detrapping of the positron, due to local thermal excitations, becomes significant. If this is indeed the case, the interpretation of DBPAS data for low prismatic dislocation loop densities

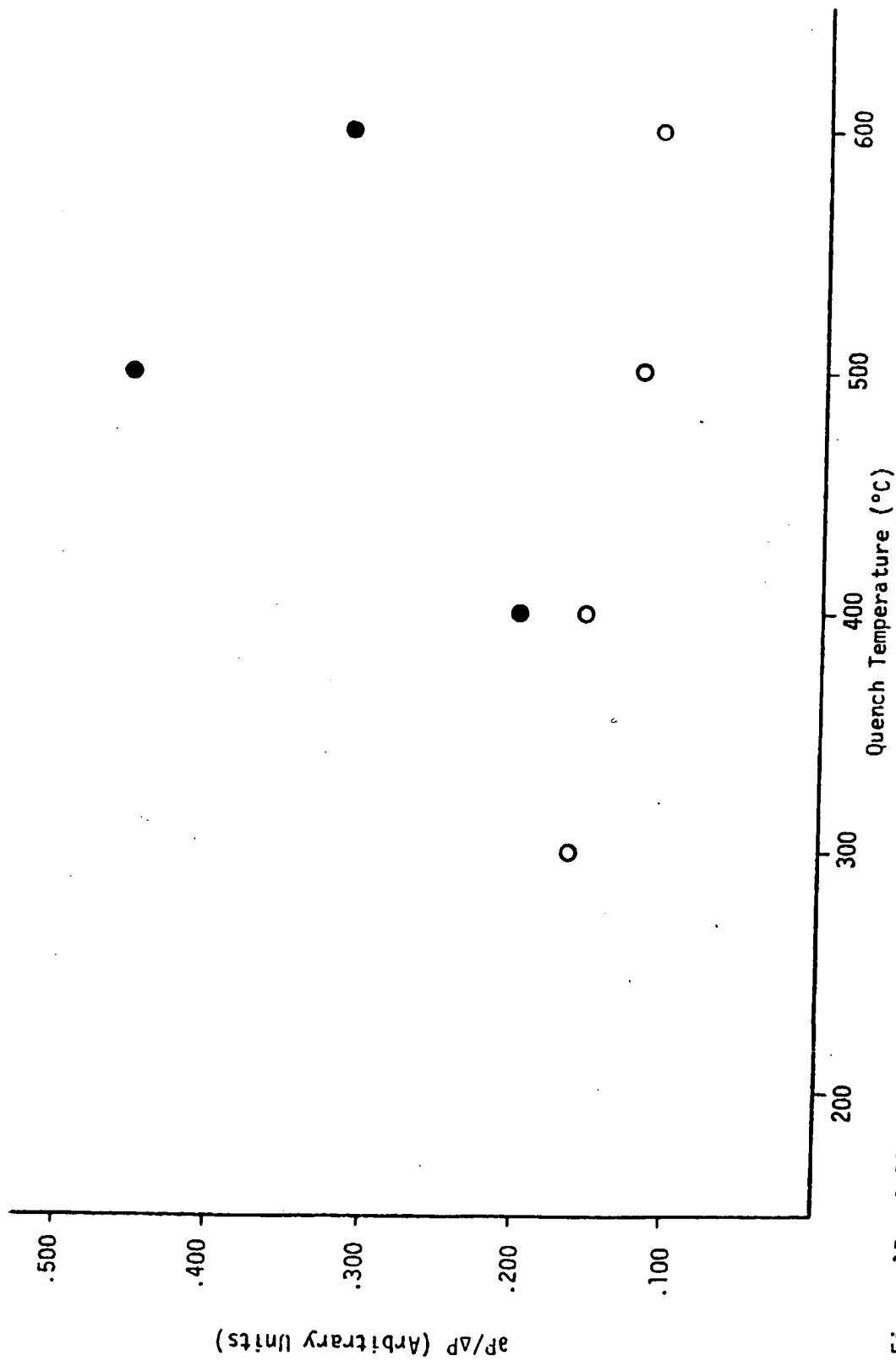


Figure 15. A Plot of the Error Parameter,  $\Delta P / \Delta P$ , for Both Dislocations (●) and Vacancies (○), as a Function of Quench Temperature for Aluminum Samples.

becomes more complicated. Clearly, additional experimental data is required to determine the exact reason for the observation that the error parameters are larger for the samples containing primarily dislocation loops than for the samples dominated by vacancy type defects.

Another interesting feature of Figure 15 is the relatively constant value of  $\delta P/\Delta P$  for vacancies. This implies that the spread in the data decreases with the quenching temperature, since clearly  $\Delta P$  decreases with  $T_Q$ . One possible explanation for this observation is related to the shift in the centroid of the experimentally obtained spectrum due to temperature and/or electronic drift. The explanation is that a centroid shift changes the peak shape of a broad and flat spectrum (low defect concentration) to a lesser extent than that of a narrow and sharp spectrum (high defect concentration). This is, of course, an extremely qualitative argument but it does predict that in a temperature/digitally controlled series of experiments the relative size of the error bars associated with the high defect concentration samples should decrease.

The preceding discussion has demonstrated the utility of deconvolution in DBPAS measurements. In summary, the use of the FFT/PS method improves the sensitivity and the relative error of the calculated lineshape parameters. In addition, deconvolution automatically corrects for a wide range of error sources which result in changes in the instrument resolution function. Deconvoluted lineshape parameters should be more directly comparable between different laboratories and, in addition, offer the potential ability to resolve fine structure in the energy spectrum which is unavailable from direct observation of the raw data. There is strong evidence (see for example Hood and Schultz, 1980) suggesting that the use of both temperature stabilization and digital stabilization, to control centroid drift, should significantly improve

the accuracy of the results.

It is interesting to note the conditions under which the undeconvoluted raw data has yielded reasonable results in the past. The main requirement is, of course, experimental conditions which yield a nearly constant resolution function,  $R(x)$ . As one would expect, temperature and digital control as well as performing the series of experiments as close together in time as possible, both tend to help yield reasonable results for the raw data. In addition, the use of "S" type parameters, which are essentially area integrals over large energy windows, will be affected only slightly by the "spreading" action of the instrumentation. The major advantages one gains by deconvolution are: 1) the ability to compare more accurately results for a wide variety of experimental conditions and, 2) the possibility of observing a more detailed structure of the positron-electron annihilation energy spectrum.

#### The Separation of Multiple Defect Types

The previous discussion has demonstrated that deconvoluted DBPAS measurements are able to detect the presence of both vacancies and dislocations in high purity aluminum. The evidence for this statement comes from controlled laboratory experiments using samples with reasonably well characterized defect distributions. In order for DBPAS measurements to become useful as an NDT tool in industrial applications it is important for the technique to be able to distinguish between multiple defect types. The reason for this requirement is that in a practical application one may require information about a particular defect type while being comparatively unconcerned with other defect types. For example, if one is interested in hydrogen embrittlement of high strength steels the "masking" of vacancies by nascent hydrogen may be of primary importance while slight variations in dislocation density may have relatively little significance.

Before discussing the results of this investigation to determine the ability of DBPAS to separate defect types a brief review of previous work in this area will be presented. The basic concept is that no two defect types will have identical electron configurations associated with them. That is, the distribution of electrons (both in terms of electron density and momentum) in the neighborhood of a single vacancy will be different than in the neighborhood of a dislocation core. Qualitatively the atmosphere will be the same, i.e. a decrease in electron density and a lower average electron momentum, but quantitatively, the measured lifetimes and fine structure of the momentum distribution should be different.

This variation in electron distribution has been verified repeatedly in the case of lifetime measurements. For example, Fluss et al. (1978) report a lifetime of  $\sim 244$  ps for positrons trapped at a vacancy in aluminum while Hautajarvi et al. (1970) give the value of  $\sim 229$  ps for a dislocation in the same material. It should be mentioned that the accurate determination of multiple lifetimes from a single experiment requires some rather involved calculations. The complexity of the procedure increases rapidly with the number of unconstrained components one attempts to resolve and, for crystalline materials, the results are questionable if more than two defect types are considered.

An alternate method of analyzing lifetime data is to calculate a single lifetime component, known as the "average" lifetime, for a sample containing multiple defect types. This approach simplifies the calculations and finds its greatest utility in experiments in which the concentration of one defect type is varying over a wide range of values while the concentrations of the other defect types remain relatively constant. Of course, such an approach results in the loss of the ability to separate defect types.

In the case of DBPAS measurements there have been fewer attempts to distinguish between different defect types. The most notable exception is the work of Mantl and Triftshauser (1978) which will be discussed in a following section. There are at least two major reasons for the lack of work in this area. The first reason is related to the comparatively poor energy resolution of the DBPAS technique. Because of the smoothing action of the instrumentation the fine structure of the energy distribution may have been hidden in previous experiments. In addition, even a slight change in the instrument resolution function during a series of experiments could mask the subtle changes in lineshape required to separate defect types. Although temperature and digital stabilization of the electronics would improve the situation, such a procedure does not address the issue of the smoothing action of the instrumentation. It is the use of deconvoluted data which offers the possibility of examining the fine structure of the electron momentum distribution.

As mentioned in the introduction, the most commonly reported Doppler lineshape parameter is the MacKenzie "S" parameter (1970). This lineshape parameter is defined as the ratio of counts in some central region of the DBPAS energy spectrum to the total number of counts in the spectrum. Since it is known that the energy spectrum becomes more sharply peaked with increasing vacancy type defect concentration it follows that the numerical value of "S" should be proportional to the total vacancy type defect concentration. The disadvantage of using this lineshape parameter is that it is unable to distinguish between multiple defect types. For example, if both monovacancies and edge dislocations are present in a sample both defect types will tend to increase the value of "S", however, it will be impossible to separate the relative defect concentrations solely from the calculated value of "S".

Mantl and Trifthauser (1978) developed a method for separating the effects of multiple defect types from experimentally obtained DBPAS data. To accomplish this they introduced a new lineshape parameter,  $I_c$ , defined as the ratio of the sum of the number of counts in two regions, (one on the low energy side and one on the high energy side of the peak) divided by the total number of counts in the spectrum. This lineshape parameter, which in this work will be referred to as the wing or "W" parameter, can be shown to decrease with increasing vacancy type defect concentration. When analyzed separately the "W" parameter suffers from the same disadvantages as the "S" parameter in that it is incapable of separating multiple defect types. However, Mantl and Trifthauser recognized that the combined use of "S" and "W" could yield useful results.

In Appendix H the derivation of their defect specific "R" parameter is recreated. To summarize, it can be shown that in theory the "R" parameter, as defined below, is concentration independent, but characteristic of the type of trapping site involved (Mantl and Trifthauser 1978). The "R" parameter is defined as:

$$R = |(S - S_B)/(W - W_B)| \quad (4.6)$$

where  $S$  is the "S" parameter for a given specimen,

$S_B$  is the value of "S" corresponding to a "defect free" specimen,

$W$  is the "W" parameter for a given specimen,

$W_B$  is the value of "W" corresponding to a "defect free" specimen.

While such a parameter is very useful, because of its ability to distinguish between multiple defect types, it still does not solve the problem completely due to some inherent theoretical limitations. The main restriction is that the analysis is valid if only one type of trapping center is present, or if one defect type is essentially dominant (Mantl and Trifthauser 1978). In

practice this means the experimenter has the ability to calculate defect type and concentration provided each sample has only one predominant type of positron trap. The difficulties arise if a sample contains two defect types, say vacancies and dislocation cores, in roughly equal concentrations.

In terms of the present experiments on high purity large grain size aluminum, because care has been taken to produce samples with only a single dominant defect type, one would expect the "R" parameter analysis to yield useful results. A family of R-type parameters can be calculated for the experiments in this work by examining ratios of the form

$$R_N = |((\Delta x(i)/\Delta x(j)))| \quad (4.7)$$

where  $R_N$  is an R-type parameter

$\Delta x(i)$  is the difference between  $x(i)$  for the current sample and the value of  $x(i)$  in a defect free sample

and  $i$  is less than  $j$

In the original work of Mantl and Trifthauser (1978) the energy windows selected to define "S" and "W" were chosen so that the annihilation events contributing to the "S" parameter corresponded to conduction (valence) electrons while core electron events resulted in contributions to the "W" parameter. This relationship restricts the possible values of  $i$  and  $j$  as defined in Equation (4.7) above.

The specific "R" parameter used in this study is defined as

$$R_1 = \left| \frac{\Delta x(1)}{\Delta(x(10) + x(11) + x(12))} \right| \quad (4.8)$$

and is defined such that the energy windows are fairly close to those used by Mantl and Trifthauser (1978). For comparison,  $R_1$  has a low energy window of ~511 - 511.32 KeV and a high energy window of ~513.88 - 514.84 KeV compared to windows of ~511 - 511.5 KeV and ~513.6 - 516.1 KeV in the referenced work.



Figure 16 is a plot of  $R_1$  as a function of quenching temperature for the background corrected raw data. Note that although samples with primarily dislocation type defects have, in general, a higher  $R_1$  value than the samples with primarily vacancy type defects, there is a significant amount of scatter in the data. The source of this scatter is not known exactly, but it is probably related to the fact that the data was collected without temperature or digital stabilization. In fact, it is plausible that the spread in  $R_1$  values is a numerical amplification of the spread in values of  $x(1)$ ,  $x(10)$ ,  $x(11)$  and  $x(12)$ . In addition to this error source, any error associated with changes in the instrument resolution function, (IRF), should also be present in Figure 16. To test the utility of deconvolution in R-parameter analysis one would like to have a data set in which the second error source is present but the first error source, related to electronic drift, is negligible. Unfortunately, such a data set can not be obtained with the instrumentation used in this study.

In an attempt to isolate the instrument resolution effects the following procedure was employed. Using the fact that all DBPAS measurements for a given metallurgical sample were performed in a comparatively short time interval (~8 hrs.), it seems reasonable to assume that the spread in calculated lineshape values is a result of the lack of temperature and digital stabilization rather than of changes in instrument resolution. This hypothesis is supported by the observation that for a given set of consecutive experiments, the IRF's measured before and after the experiments showed only very slight changes. If this assumption is true, one may be able to artificially select a subset of the original data which qualitatively corresponds to a series of experiments with reduced electronic drift error. In this work each group of DBPAS measurements, corresponding to a sample subjected to

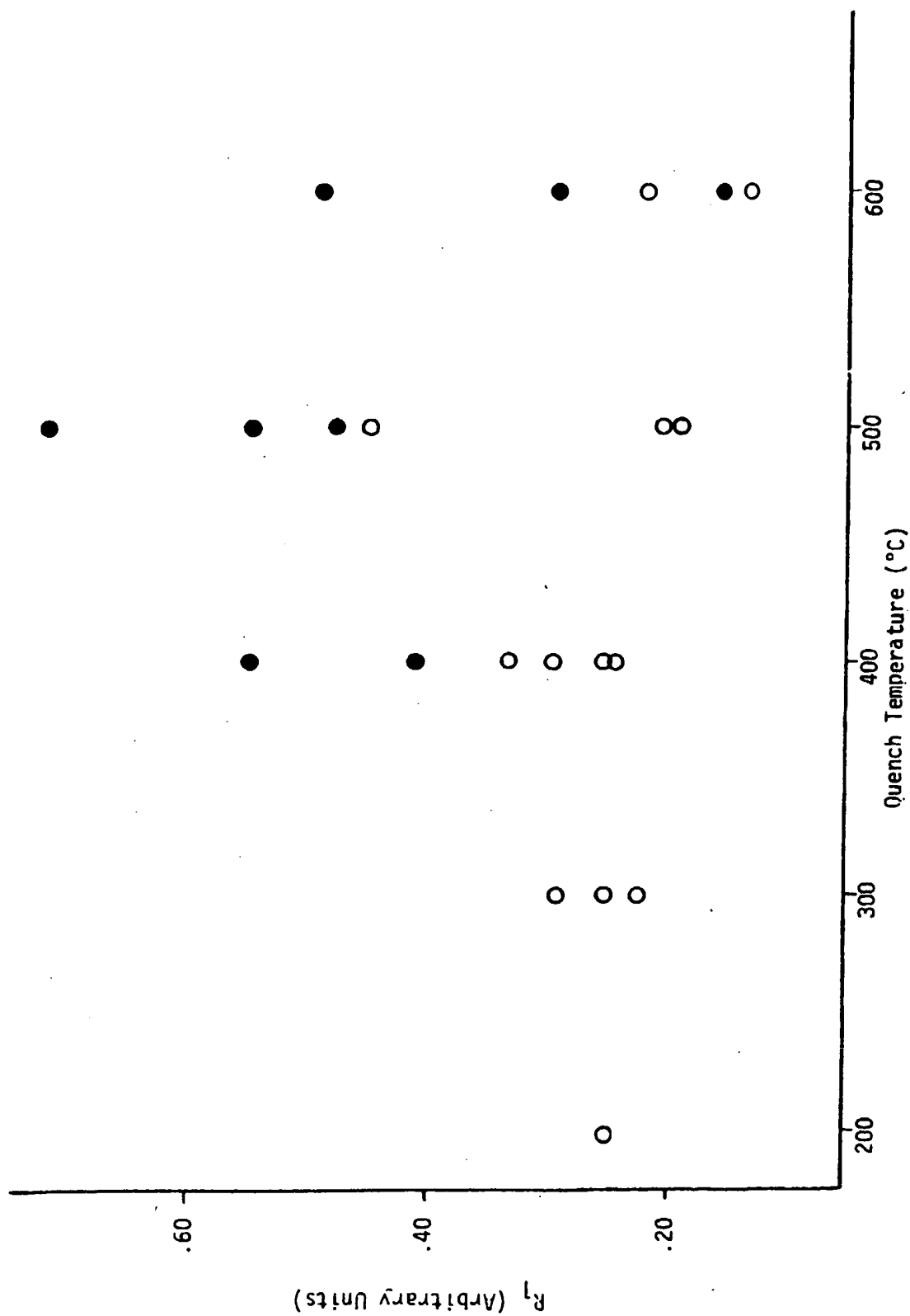


Figure 16. A Plot of  $R_1$  Values for the Raw Data vs. Quench Temperature for Dislocations (●) and Vacancies (o).

a given thermal treatment, was independently examined and an average value for each relevant lineshape parameter was calculated. The corresponding  $R_1$  values calculated from the average lineshape parameter values are shown in Figure 17. These values are, as in Figure 16, for the background corrected raw data.

A similar procedure was then employed for the deconvoluted data. That is, average  $x(i)$  values were calculated for each specimen and then used to calculate  $R_1$  values. These  $R_1$  values for the deconvoluted data are shown in Figure 18. It is quite apparent that the spread in  $R$ -parameter values has been increased by the FFT/PS deconvolution algorithm. The explanation for the increased scatter in the data is not immediately obvious. In a previous section of this work it was demonstrated that deconvolution improved the quality of the calculated  $x(1)$  lineshape parameter values. That is, the spread in  $x(1)$  values for DBPAS measurements performed on a given sample was decreased (refer to Table 1). One could argue that decreasing the scatter of  $x(1)$  for a given sample should improve, or at least not adversely affect, the amount of scatter in the calculated  $R_1$  values. The difficulty is, of course, that  $R_1$  depends not only on  $x(1)$  but also on the values of the parameters  $x(10)$ ,  $x(11)$ , and  $x(12)$ . These parameters are defined in the tail regions of the energy distribution and it is in this region that one must look to understand the difficulty associated with the calculation of  $R$ -type parameters for FFT/PS deconvoluted data.

Figures 19 and 20 are a comparison of the energy distribution (raw data) for a sample quenched from 600°C and then subsequently annealed at 523°K. Figure 19 shows the differences in the peak region of the spectra. Recall that  $x(1)$  is defined over the energy window of 511 - 511.32 KeV. In Figure 20 the vertical scale has been expanded so that the differences in the tail region are visible. Several important features of the energy

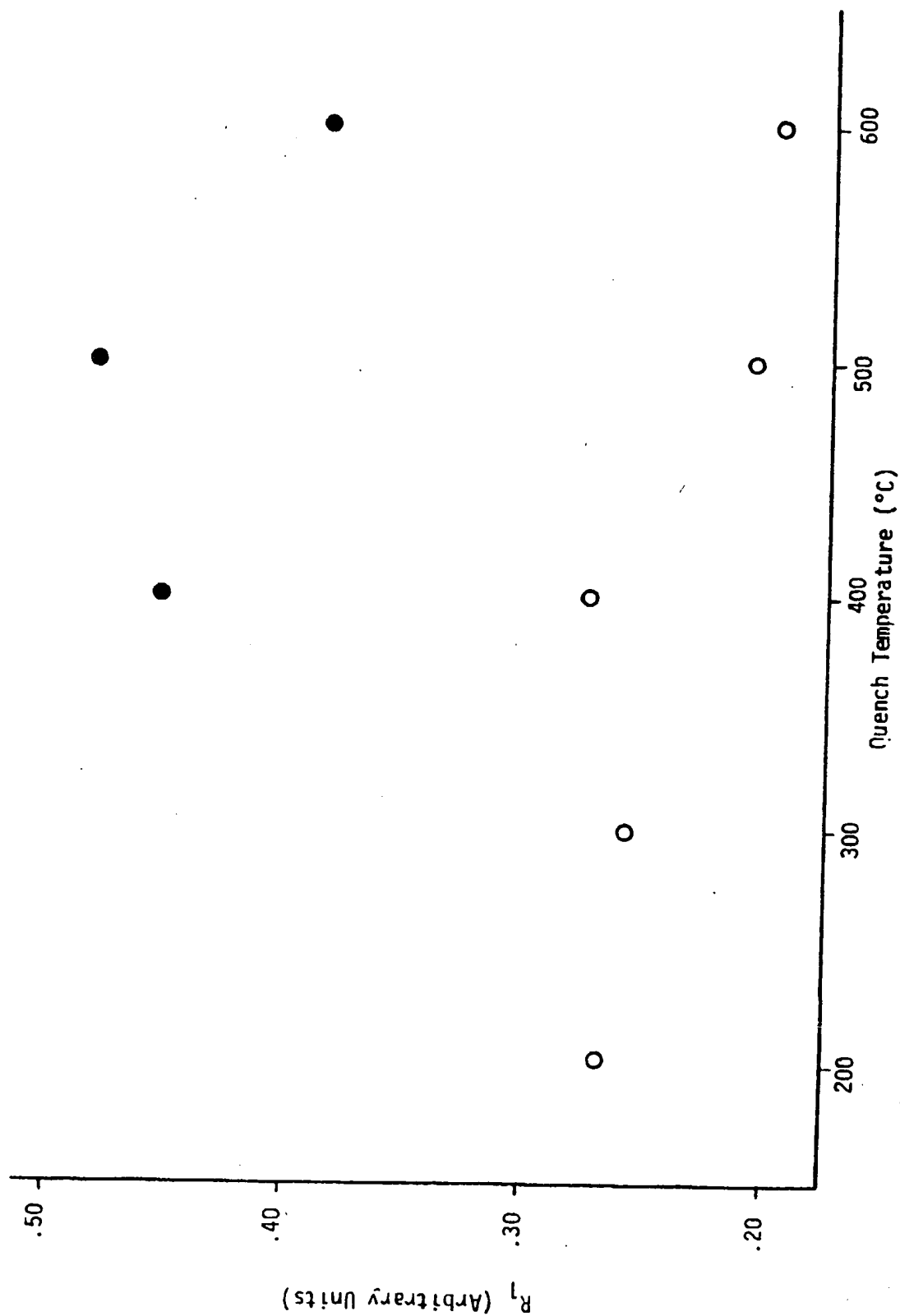


Figure 17. A Plot of  $R_1$  Values as Calculated from Average Lineshape Parameter Values of the Raw Data vs. Quench Temperature for Dislocations (●) and Vacancies (○).

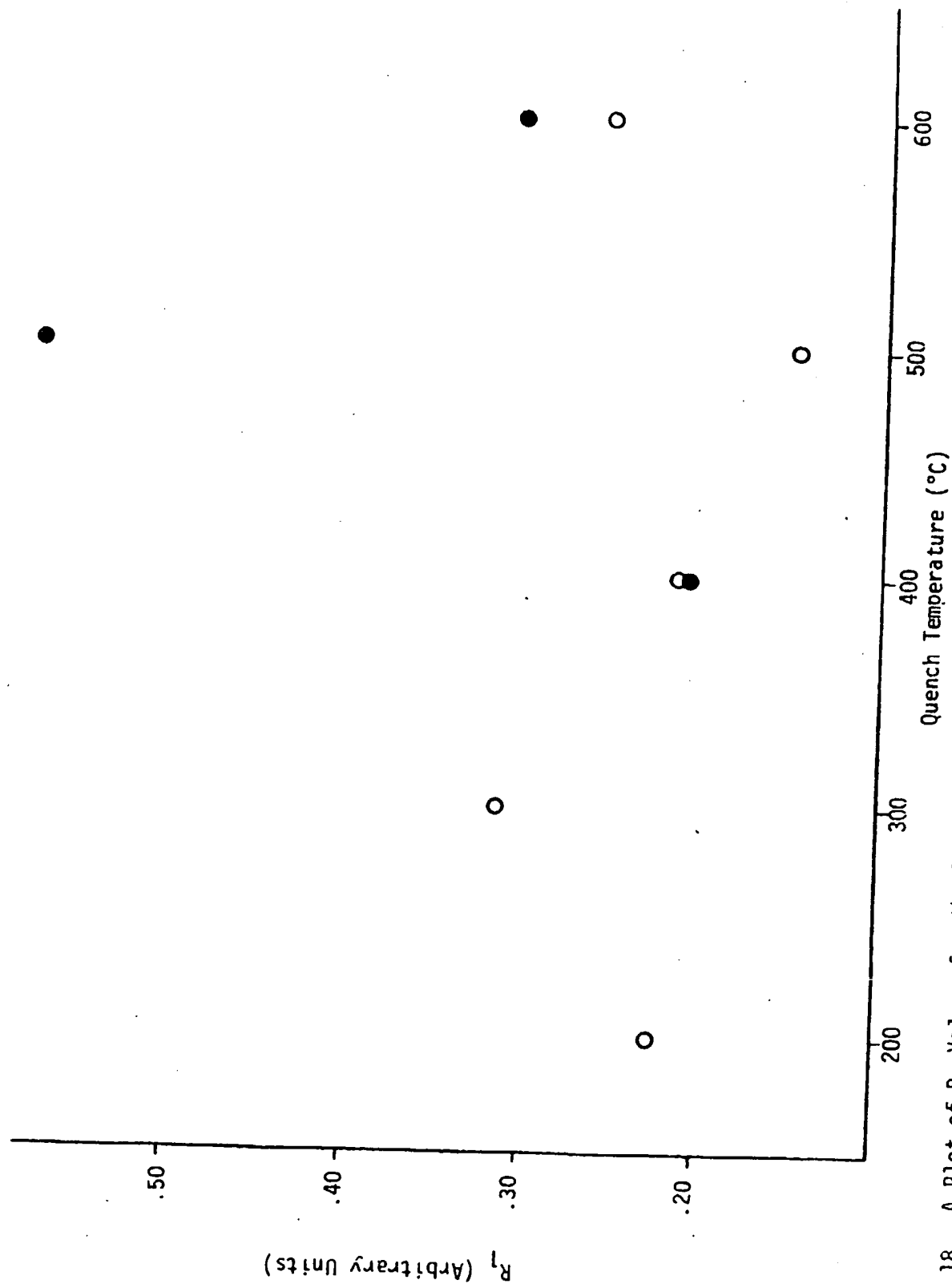


Figure 18. A Plot of  $R_1$  Values for the Deconvoluted Data, Using all Retained Coefficients, vs. Quench Temperature.

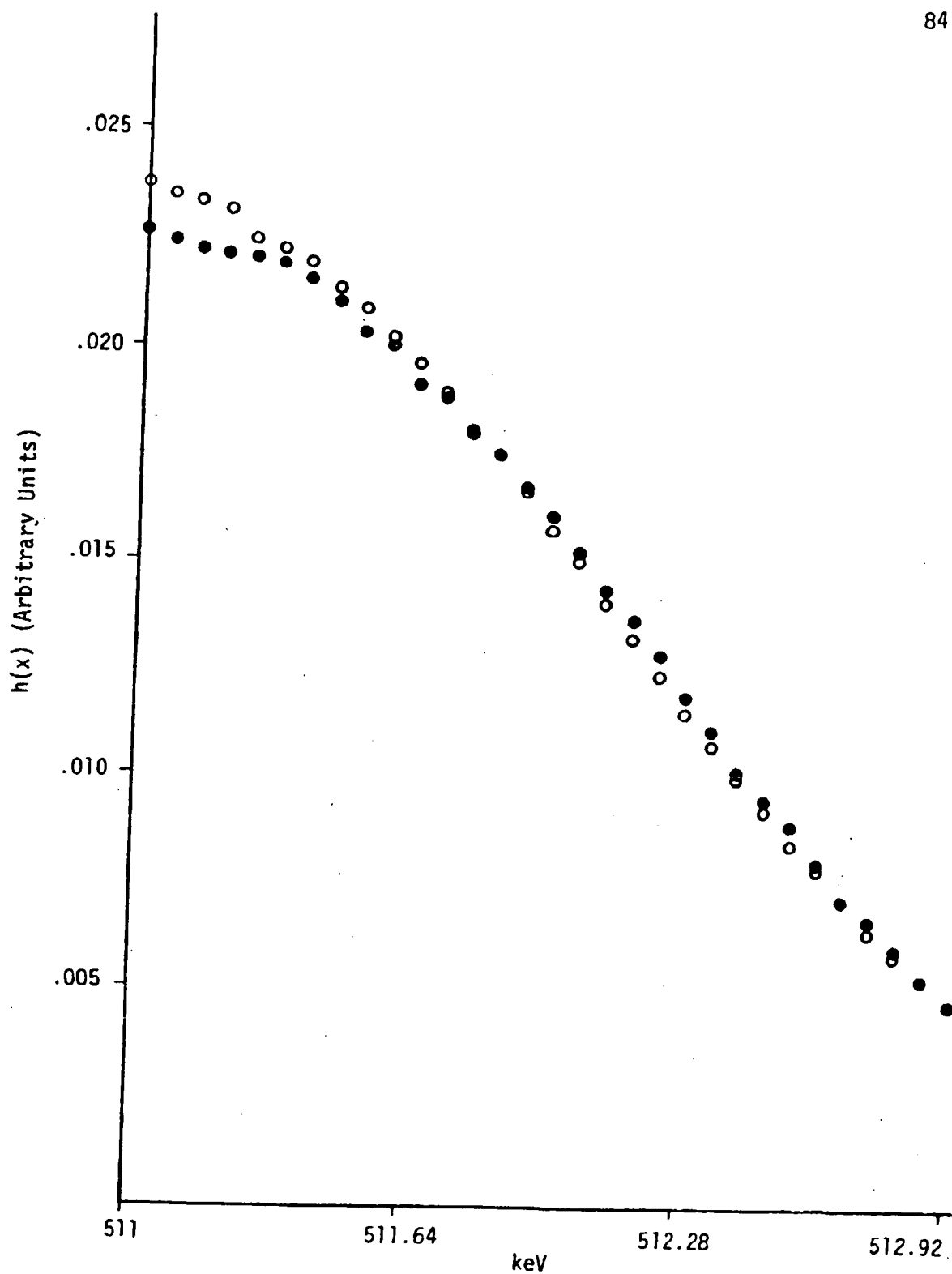


Figure 19. A Comparison of the Raw Data for an Aluminum Sample Quenched from 600°C (o) and the Same Sample After the High-Temperature Anneal (•).

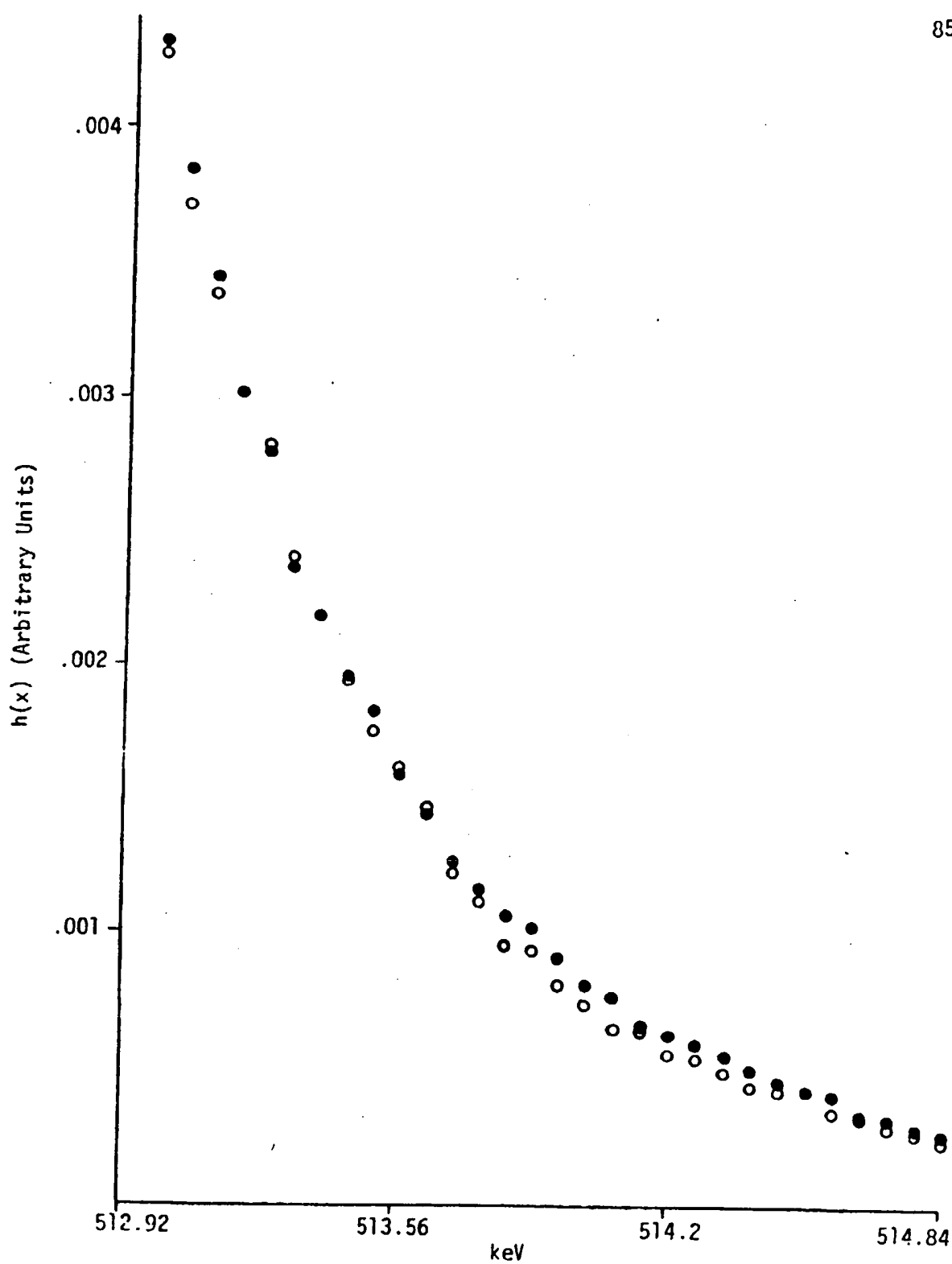


Figure 20. A Comparison of the Tail Region of the Raw Data for an Aluminum Sample Quenched from 600°C (o) and the Same Sample After the High Temperature Anneal (•).

distribution in the tail region are apparent. First, it must be noted that in general the data points for the quenched sample lie below their counterparts for the annealed sample as predicted by theory. Secondly, the statistical, or counting, error from channel to channel can be recognized. The former observation is believed to account for the qualitative agreement between the theoretical predictions and the calculated  $R_1$  values for the raw data. The latter observation, statistical noise, is the most probable explanation (along with electronic noise) for the spread in  $R_1$  values as shown in Figure 16.

For comparison, Figures 21 and 22 show the change in energy distribution for the same two samples after deconvolution using the FFT/PS algorithm. In Figure 21, which shows the peak region, some of the desirable features of deconvolution are evident. Most notably, the statistical channel to channel noise has been removed (compare with Figure 19). A careful comparison of the raw and deconvoluted data shows that deconvolution does indeed decrease the full width half maximum as discussed previously.

Figure 22 shows, with an expanded vertical scale, an undesirable feature of the FFT/PS deconvolution method. It was mentioned in Chapter III that truncating the Fourier coefficients is equivalent to the insertion of a low pass filter and it is well known that low pass filtering introduces certain errors. Recalling that the Fourier analysis of the convolution equation, neglecting the statistical counting error, yields the following equation for the desired function  $f(x)$ :

$$f(x) = \int_{-\infty}^{+\infty} u(w)F(w)e^{-iwx}dw \quad (4.10)$$

where  $u(w) = 1$  for  $w \leq w_c$   
 $= 0$  for  $w > w_c$



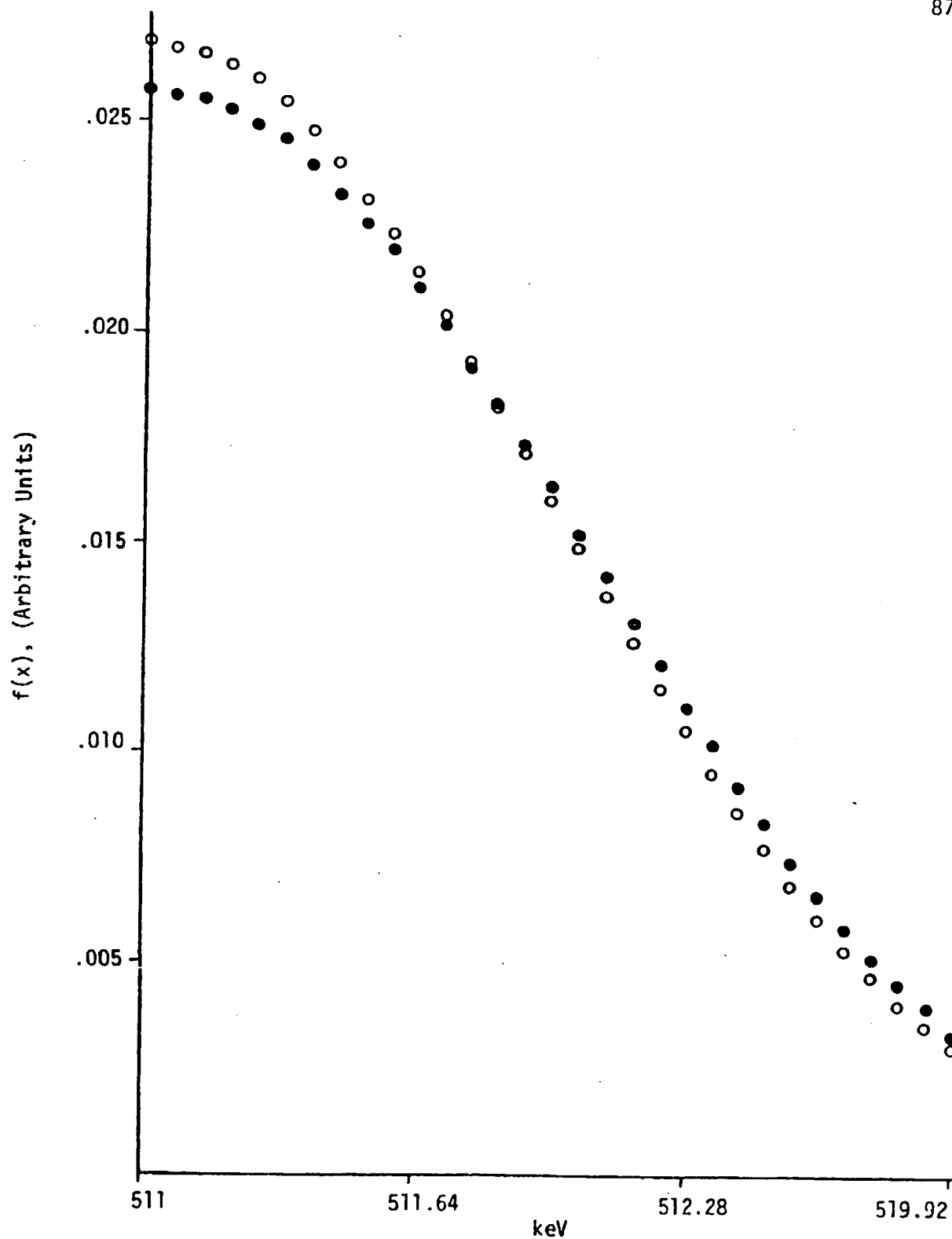


Figure 21. A Comparison of the Deconvoluted Data for an Aluminum Sample Quenched from  $-600^{\circ}\text{C}$  (o) and the Same Sample After the High Temperature Anneal (●).

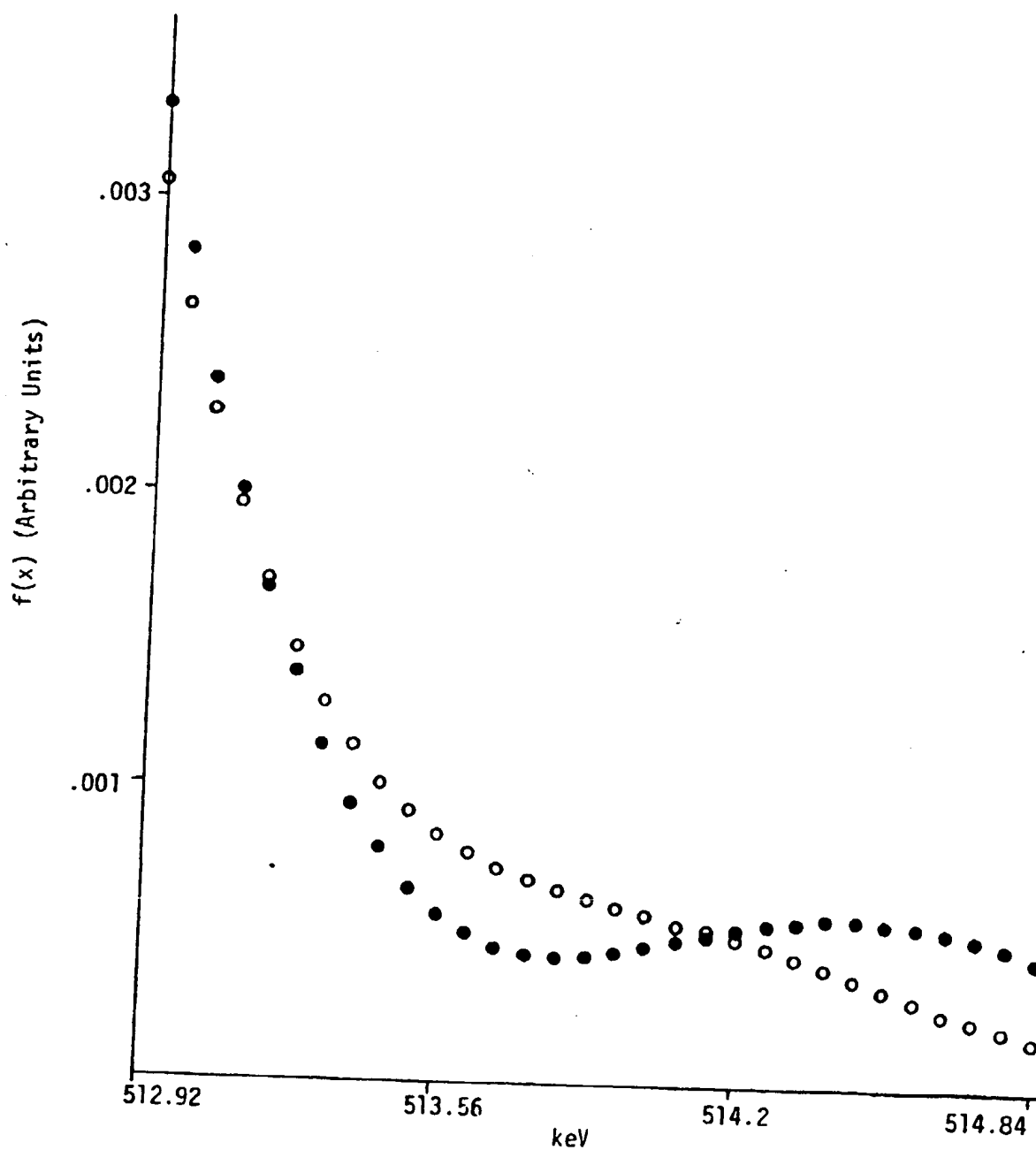


Figure 22. A Comparison of the Tail Region of the Deconvoluted Data for an Aluminum Sample Quenched from 600°C (o) and the Same Sample After the High Temperature Anneal (•).

and  $w_c$  corresponds to the frequency which is the first minimum of the power spectrum of  $h(x)$

It can be shown (Bendat and Piersol 1971) that error introduced by such a filter is proportional to  $(1/w_c)\sin(2\pi x w_c)$ . The significance of this result is that the amplitude of the error is inversely proportional to  $w_c$  (the number of retained coefficients), and the frequency of the resulting error is proportional to  $w_c$ .

This effect is clearly evident in Figure 22. The open circles represent the annealed sample for which the power spectrum analysis yielded a  $w_c$  of fifty six (56) coefficients. For comparison, the filled circles, corresponding to the quenched specimen are calculated using sixty seven (67) retained coefficients. It is quite obvious that the error amplitude is decreased for the sample with the larger number of retained coefficients. A careful analysis of the data shows that the frequency of the error is proportional to  $w_c$ .

With this information, it is now possible to explain the increased scatter in  $R_1$  values displayed by the FFT/PS deconvoluted data. Because the energy window used to define the wing parameter (513.88 - 514.84 KeV) is in the range where the amplitude of the filtering error is of the same order of magnitude as the amplitude of the true signal, this error source is significant. If the wavelength of the error source was small compared to the size of the energy window, then the error contribution would average to nearly zero and this difficulty would not exist. The problem arises because the energy window spans less than a period of the sinusoidal error source and it spans a different fraction of the period depending on the number of retained Fourier coefficients. For example, in Figure 22 the energy window of interest for the annealed sample spans approximately the positive half cycle of the

sinusoidal filtering error. This leads to a calculated wing parameter with an artificially high value, which in turn distorts the value of the calculated  $R_1$  parameter. Since multiple DBPAS measurements on even the same metallurgical sample may yield slightly different numbers of retained coefficients, this error source can lead to a scatter of  $R_1$  parameters for a single sample. Because the number of retained coefficients varies more widely between samples of differing defect density (or defect structure), the spread in  $R_1$  values becomes larger for such samples.

Having related the scatter in  $R_1$  values for the deconvoluted data to the phase and amplitude of the filtering error, it is now possible to try to correct for this error source. There are at least three ways to proceed. First, one could simply extend the experimental collection times to get a higher number of total integrated counts in the energy spectrum. This would result in better counting statistics, and a higher number of retained coefficients (Schaffer *et al.* 1984). Since an increased number of retained coefficients decreases the amplitude of the filtering error, such a procedure should yield better  $R_1$  values. Unfortunately one must pay a very high price in terms of collection time to significantly increase  $w_c$ .

Another approach is to use the same number of retained coefficients for each data set. That is, one could calculate a  $w_c$  value for each data set to be analyzed and then deconvolute all of the data sets using the minimum value of  $w_c$ . Using this procedure will not eliminate the error source, but it will be controlled so that its contribution to the calculated values of  $R_1$  does not vary between experiments. Clearly such an approach has several inherent disadvantages. The primary disadvantage is that the absolute values of  $R_1$  will be a function of the particular value of  $w_c$  used in the deconvolution procedure. One would prefer an approach which eliminates, or at least

greatly reduces, the filtering error rather than simply controlling it. A method for accomplishing this goal will be discussed in the final chapter of this work. What follows here is a discussion of controlling the error source in the tail region by using the same number of retained coefficients for each data set.

Figure 23 shows the energy window used to calculate the wing parameter. The vertical scale has been greatly expanded to show the fine detail of the structure. This plot is for the same samples as in Figure 22, but in this case both data sets have been deconvoluted using fifty seven (57) coefficients. As expected, the filtering error is in phase. In addition, as theory predicts, the data points corresponding to the quenched sample are below the points representing the annealed sample. One can extend this procedure to all of the other data sets and then calculate the corresponding  $R_1$  parameters in the manner previously described. The results are presented in Figure 24. When this figure is compared with Figure 18 it can be seen that the filtering error does influence the scatter in  $R_1$  values. By simply controlling this error source, i.e. by using the same number of retained coefficients in the deconvolution of all data sets, the quality of the calculated  $R_1$  values can be improved.

To summarize this section, the average values of  $R_1$ , along with the associated standard deviations for each of the two defect types, have been calculated for four different types of data. The four data types are: 1) the entire raw data set (Fig. 16), 2) the average raw data (Fig. 17), 3) the averages of the deconvoluted data as obtained directly from the FFT/PS algorithm (Fig. 18), and 4) the averages of the deconvoluted data using the same number of retained coefficients for each data set (Fig. 24). These values are shown in Table II.

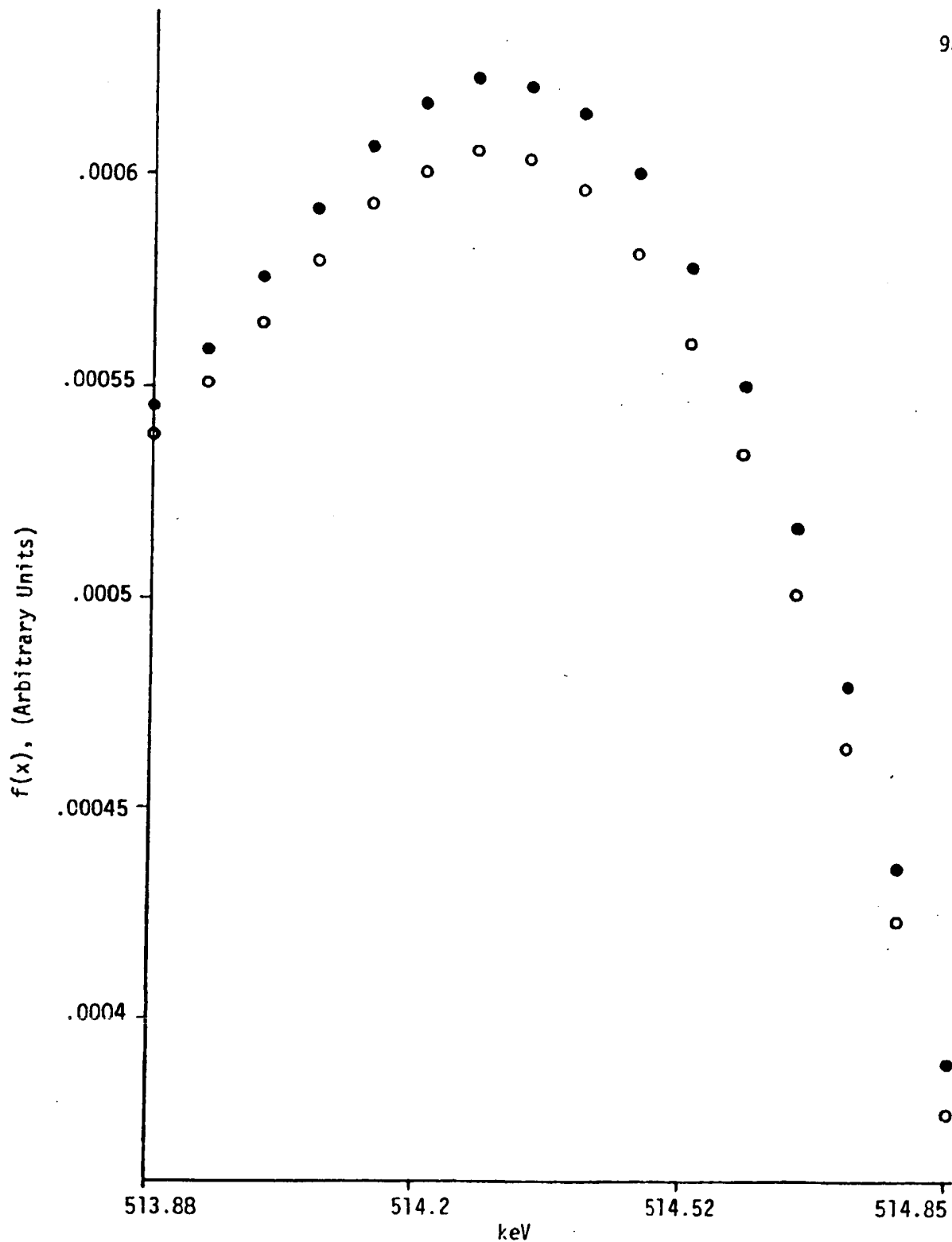


Figure 23. A Comparison of the Tail Region of the Same Samples as in Figure 22. In this Plot Both Data Sets have Been Deconvoluted Using the Same Number (57) of Reconstruction Coefficients.

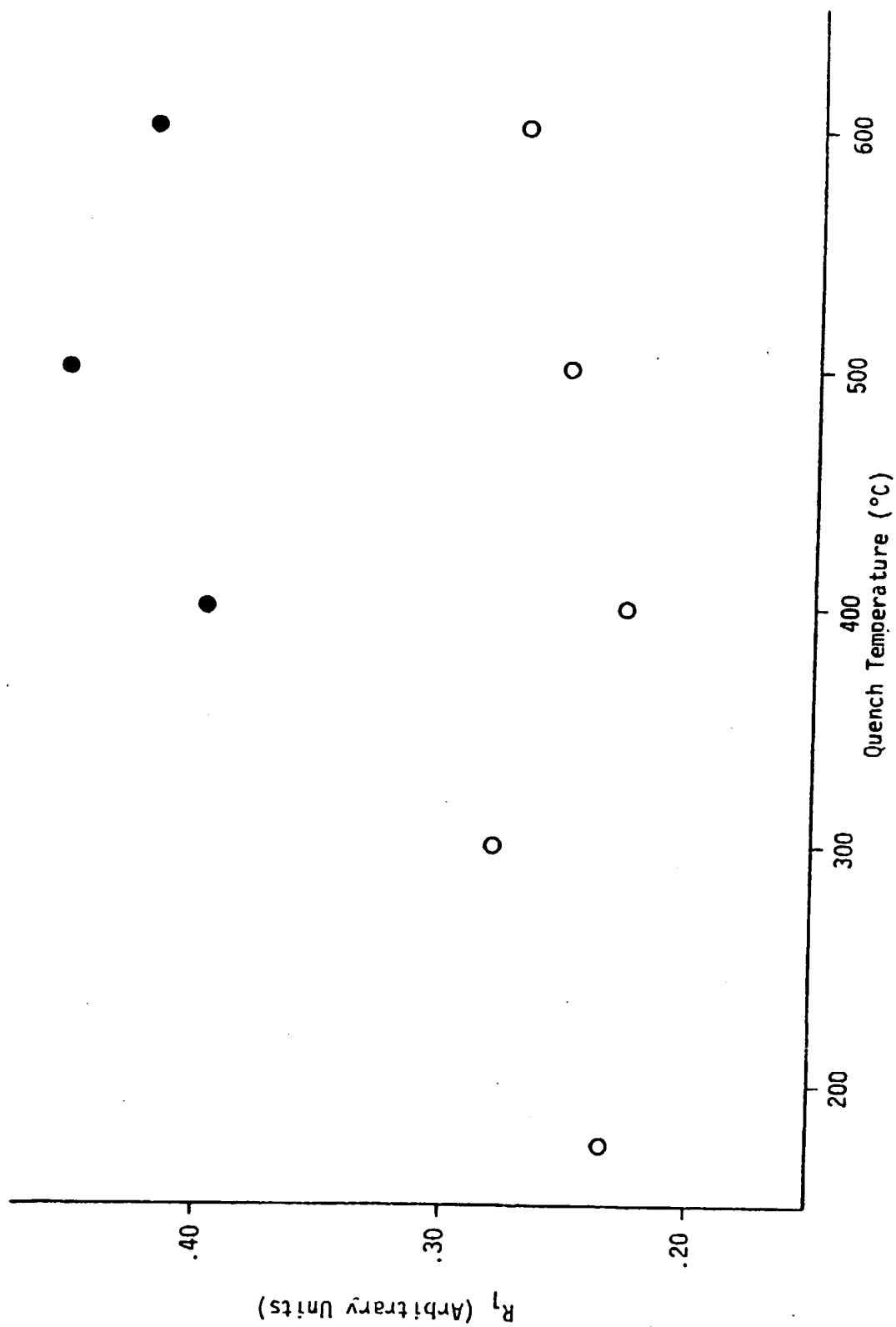


Figure 24. A Plot of  $R_l$  Values, for the Deconvoluted Data Using a Constant Number of Reconstruction Coefficients, Comparing Dislocations (●) with Vacancies (○).

Table II

Data Type	$R_{avg}(Vac.)$	$R_{avg}(Dis.)$	$\sigma(Vac.)$	$\sigma(Dis.)$
1	.261	.458	.0677	.1751
2	.240	.440	.0347	.0492
3	.234	.364	.1515	.1876
4	.253	.424	.0216	.0290

Note that the largest standard deviations occur for the deconvoluted data when the maximum number of coefficients are used in the reconstruction of each  $f(x)$ . This result is explainable in view of the previously discussed relationship between  $w_c$  and the amplitude and phase of the filtering error. When this error source is controlled by using a single value of  $w_c$  for the deconvolution of all data sets (data type 4) the calculated standard deviations are improved. With this modification to the FFT/PS deconvolution algorithm it appears that if one defect type dominates, one can distinguish between vacancies and prismatic dislocation loops, in high purity large grain size aluminum.

To conclude the results and discussion chapter of this work the even central moments of the annihilation energy distributions will be discussed. As mentioned in the introduction, because this technique has the potential to distinguish between multiple defect types, this family of lineshape parameters is interesting from a theoretical point of view. Briefly, this possibility arises because the higher order central moments are increasingly dominated by the tail regions of the energy distribution. Thus, the low moments give information about the central region (S-type information) while the higher order moments yield W-type information. Unfortunately, this analysis is extremely sensitive to small errors in the tail regions which are greatly



amplified by moment analysis. Therefore the statistical error ruled out moment analysis in the raw data while filtering error precluded its use in the deconvoluted data. It is possible, however, that an improved filtering technique, discussed in the following chapter, will allow moment analysis to become a useful tool in future work.

## CHAPTER V

### CONCLUSIONS AND FUTURE WORK

Over the past several decades positron annihilation spectroscopy has evolved into an accepted tool for the study of the defect structure of materials. Of the three most common measurement techniques Doppler broadening offers the advantages of reasonably high data collection rates, comparatively low source strength requirements, and few geometrical experimental constraints. It is therefore the technique with the best chance of making the transition from a laboratory research device to an industrial NDT tool. Unfortunately, DBPAS measurements suffer from some inherent limitations. The two most significant limitations are comparatively poor instrumental energy resolution and limited ability to distinguish between multiple defect types. Based on these observations, the two primary goals of this work were: 1) to develop an efficient, model independent, noniterative deconvolution method, and 2) to improve the ability of DBPAS to distinguish between multiple defect types.

In order to improve the effective energy resolution of the DBPAS instrumentation the fast Fourier transform/power spectrum analysis deconvolution algorithm was developed (Schaffer et al. 1984). The convolution integral equation was shown in Chapter III to describe the relationship between the experimentally observed energy distribution, the instrument resolution function,

and the "true" or desired Doppler broadened energy distribution. Fourier analysis was then used to obtain what appeared to be a closed form solution for the desired function. Unfortunately, it was demonstrated that this solution is not unique. The statistical nature of the experiment places the equation into the mathematical class of incorrectly posed problems. In an attempt to find a reasonable approximation to the desired function the dispersion of the apparent closed form solution was investigated. It was observed that the dispersion became unbounded if the power spectrum of the statistical experimental "noise" remained finite for high frequencies. That is, the mathematical difficulties are related to the "white noise" component present in the experimental data.

Since the shape of the power spectrum of the raw data appeared to be of critical importance, particularly if it contained a "white" component, it was investigated in greater detail. From this analysis it was observed that the overall signal-to-noise ratio for the experiment was very high, being of order  $\sim 10^6$ . Using the observation that the power spectrum falls steeply to a minimum before rising due to the white noise component in the experimental data, the FFT/PS method developed in this work truncates the Fourier coefficients of the raw data at this first minimum. The logic behind this approach is rather simple. Retaining fewer coefficients than this implies a smoothing of the deconvoluted spectrum and the loss of potentially valuable structural information. Adding more coefficients gains nothing because the signal-to-noise ratio is of order one at the minimum. In addition, truncating the coefficients at the point where the white noise begins to dominate forces the dispersion of the solution to remain finite.

The truncated Fourier transform of the raw data is then used to calculate a reasonable estimate of the desired function. It was shown in Chapter

III that this deconvolution algorithm does indeed yield reasonable results. It is well known that truncating the coefficients in the manner described results in the introduction of a filtering error. The consequences of the introduction of this error source were discussed in detail in Chapter IV and will be reviewed below.

In order to test the utility of the FFT/PS deconvolution algorithm a series of high purity, large grain size aluminum samples, containing known defect types and concentrations, were prepared. The thermal treatments employed to prepare the samples containing various concentrations of monovacancy or dislocation type defects were described in detail in Chapter II. Basically the procedure consisted of a two stage quench from elevated temperatures followed by a low temperature anneal and finally a high temperature anneal. The DBPAS spectrum was measured after each of these three thermal treatments. It was assumed, without transmission electron microscopy but based on arguments presented in Chapter II, that each of the three treatments introduced a single dominant defect type. These defect types are: 1) vacancies, after the quench, 2) prismatic dislocation loops, after the low temperature anneal, and 3) the bulk or "defect free" material, after the high temperature anneal.

In Chapter IV the lineshape parameters of the DBPAS measurements were calculated (for both the raw and the deconvoluted data) as a function of quenching temperature. A comparison of these lineshape parameters revealed several interesting trends. First and foremost, it appears that the FFT/PS deconvolution method is capable of automatically correcting for accidental and/or intentional changes in the instrument energy resolution function (IRF). It must be mentioned that there are some restrictions on the types of changes in the IRF which deconvolution can correct and these restrictions are discussed in detail in Chapter IV. Another advantage of the use of deconvoluted

data is that results obtained in different laboratories should be more directly comparable if the instrumental broadening effects have been accurately removed. This ability to directly compare numerical values for standard lineshape parameters should help to clear up some of the discrepancies which currently exist in the literature.

While it is fairly clear that accurate deconvolution should correct for changes in the IRF and improve the agreement of results obtained using different instrumentation, some of the other trends observed from the comparison of lineshape parameter calculated before and after deconvolution were not entirely predictable. For example, based on the limited results presented in this work, both the sensitivity and the accuracy of the lineshape parameters are improved by deconvolution. One explanation for the improved accuracy, i.e. the decrease in the error ratio, of the deconvoluted lineshape parameters is that the filtering operation has essentially removed the statistical counting errors present in the raw data. A comparison of multiple DBPAS measurements performed on the same sample show that before deconvolution, differences in the statistical counting error in the peak region can lead to a spread in calculated  $x(1)$  values. However, after deconvolution, the energy distributions show less variation from experiment to experiments, and hence yield  $x(1)$  values with correspondingly less scatter.

The increased sensitivity of the deconvoluted parameters may be related to the removal of the "broadening" action of the experimental instrumentation. Recall that the major effect of the instrumentation is to spread counts from one channel into several adjacent channels. Therefore, the true lineshape changes in the annihilation energy distribution, which are caused by the trapping of positrons at defects, are spread out over a much larger number of channels in the raw data. The deconvolution procedure corrects for

this effect and, in so doing, restores some of the details of the "true" annihilation energy distribution. This results in an increase in observed defect sensitivity.

Even with the improvements in lineshape sensitivity and accuracy due to the FFT/PS algorithm, the DBPAS technique must have the additional ability to distinguish between multiple defect types if it is to become a useful tool in the study of industrial materials. In order to investigate the usefulness of deconvolution in resolving multiple defect types, a comparison was made between the values of the  $R_1$  parameters calculated before and after deconvolution. If the number of reconstruction Fourier coefficients is allowed to vary between samples, as would be the case if the FFT/PS method is used in its standard form, then the calculated  $R_1$  parameter value yields little useful information.

In order to explain this somewhat negative result, the consequences of truncating the Fourier coefficient, also known as low pass filtering, were examined in greater detail. It is known that the truncation of the coefficients introduces a filtering error with an amplitude inversely proportional to the number of retained coefficients,  $w_c$ , and a frequency which is proportional to  $w_c$ . Ideally, one would like to greatly increase the number of retained coefficients in order to reduce the amplitude of this filtering error. Unfortunately, as  $w_c$  is increased beyond the first minimum of the power spectrum the calculated solution will contain large, rapidly oscillating components which are not physically plausible. The mathematics of this argument were presented in detail in Chapter III.

Since it is not possible to reduce the amplitude of the filtering error by arbitrarily increasing  $w_c$  an alternative method was developed to improve the calculated values of  $R_1$  for deconvoluted data. The first step in

the procedure is to calculate a  $w_c$  value for each of the data sets to be analyzed. Next the minimum value of  $w_c$  is selected and subsequently employed in the deconvolution of all the data sets. Using this procedure does not eliminate the filtering error, but rather controls the source such that its contribution to the calculated  $R_1$  values remains approximately constant from experiment to experiment. Although this solution is not ideal, the data presented in Chapter IV clearly indicate that by controlling the error source the reproducibility of the  $R_1$  parameter, and its usefulness in the separation of multiple defect types, is improved.

To summarize this section it can be stated that the FFT/PS deconvolution algorithm appears to improve the quality of the data as obtained from DBPAS measurements in several ways. These improvements, as presented in this work include: 1) automatic correction for accidental and/or intentional changes in the instrument resolution function, 2) the potential of direct comparison of quantitative results between laboratories, 3) an improvement in both lineshape sensitivity and accuracy, and 4) the ability to distinguish between vacancy and dislocation type defects, as demonstrated for large grained high purity aluminum.

To generalize these conclusions, further studies of this kind should be performed using a variety of different alloys with known defect type and concentration. In addition, it would be extremely useful to repeat the entire series of experiments using a DBPAS system which is both digitally and temperature stabilized. Such a data set would allow one to separate the effects of instrument broadening, which are theoretically correctable by deconvolution, from the nonsystematic effects of instrumental drift. In addition, spectra collected on a stabilized system could conceivably possess better counting statistics (due to longer allowable experimental times) which could in turn

increase the number of coefficients retained in the deconvolution procedure.

Additional experiments should be performed on the FFT/PS algorithm. These experiments could be designed to investigate alternative filtering schemes with the hope of reducing the associated filtering error. Most notably, the use of cosine tapering appears to be a viable approach. Once the filtering error has been reduced or eliminated the utility of moment analysis in the separation of multiple defect types should be reinvestigated. It is not known at this time if the current FFT/PS algorithm is capable of distinguishing the formation of positronium in a nonmetallic material. Positronium would show itself as a relatively high frequency characteristic of the annihilation energy distribution. The key factor is determining whether the frequencies required to observe positronium are to the left of the first minimum in the power spectrum. This question could be investigated using both numerically simulated data and data obtained from materials in which positronium is known to form.

In order for Doppler broadened positron annihilation spectroscopy to become a widely used industrial NDT tool it must continue to evolve in several ways. Its energy resolution must improve, the ability to separate multiple defect types must be further developed, and the reproducibility and reliability of the experimental results must increase. Although the fast Fourier transform/power spectrum analysis deconvolution technique has clearly not solved all of these problems, the results presented in this work indicate that this method measurably improves the quality and reproducibility of the information obtainable from DBPAS measurements, and serves as a sound basis for future refinements.



## APPENDICES

## APPENDIX A

In this appendix the equations used to theoretically determine the stopping, or thermalization, times of positrons in metals will be discussed. For a more complete treatment of this subject see Brandt and Arista (1982). As briefly mentioned in the text, positrons slow down mainly by two mechanisms: 1) ionization and excitation of electrons which is characterized by the electronic stopping power,  $S_e$ , of the material, and 2) phonon interactions characterized by the phonon stopping power,  $S_{pH}$ . The second mechanism dominates at epithermal energies below some critical value  $E_c$  characteristic of the material. In the range from the maximum source energy,  $E_s$ , to  $E_c$  the electronic stopping power dominates. This energy range can be further divided into two subranges separated by the photon energy for which the material has its maximum stopping power,  $E_m$ . In the range  $E_m < E < E_s$  an accurate estimate for the stopping times is given by

$$t_1 = 7.7\rho^{-1}(E_i/mc^2)^{1.2} \quad (A.1)$$

where  $t_1$  is the stopping time in psec

$\rho$  is the density of the material in  $\text{g/cm}^3$

$mc^2$  is 0.511 MeV

and  $E_i$  is the initial positron energy

since

$$E_i/mc^2 < E_s/mc^2 - 1 \quad (A.2)$$

we can rewrite Eq. (A.1) as

$$t_1 \sim 7.7\rho^{-1}$$

(A.3)

which for most metals of interest results in stopping times of  $\sim 1$  psec for the high energy region.

For  $E_c < E < E_m$  the electron gas theory of conduction electrons with Fermi energy  $E_f$  is used to produce the equation

$$S_e = \frac{\pi}{105} \frac{m^2}{h} \left[\frac{2}{m}\right]^{3/2} \frac{E^{5/2}}{E_f} \quad (\text{A.4})$$

in the case of a continuous slow down approximation

$$t_s = - \left[\frac{m}{2}\right]^{1/2} \int_{E_i}^{E_f} \frac{dE}{E^{1/2} S} \quad (\text{A.5})$$

Substituting Eq. (A.4) into Eq. (A.5) and integrating yields

$$t_2 = \frac{105}{8\pi} \frac{h E_f}{E_f^2} \quad (\text{A.6})$$

where  $t_2$  is in picoseconds.

In the low energy region  $3/2 KT < E < E_c$  phonon interactions dominate with

$$S_{PH} = \frac{2}{\pi} \frac{m^3 w^2}{h^4 d} E \quad (\text{A.7})$$

where  $w$  is the lattice deformation potential.

Eq. (A.4) can be combined with Eq. (A.7) to define  $E_c$  yielding

$$E_c^{3/2} = \frac{105b^2}{2^{1/2}\pi^2} \frac{m^{5/2} E_f^3}{h^3 \rho} \quad (\text{A.8})$$

Inserting Eq. (A.7) into Eq. (A.5) and integrating from  $3/2 KT$  to  $E_c$  as defined by Eq. (A.8) yields

$$t_3 = \frac{\pi h^4 \rho}{3^{1/2} w^2 (K_B T)^{1/2}} \left[ 1 - \left( \frac{3K_B T}{2E_c} \right)^{1/2} \right] \quad (\text{A.9})$$

using typical values for common metals it can be shown that

$$t_3 > t_2 > t_1 \quad (\text{A.10})$$

Thus, phonon thermalization times are larger than electronic stopping times.

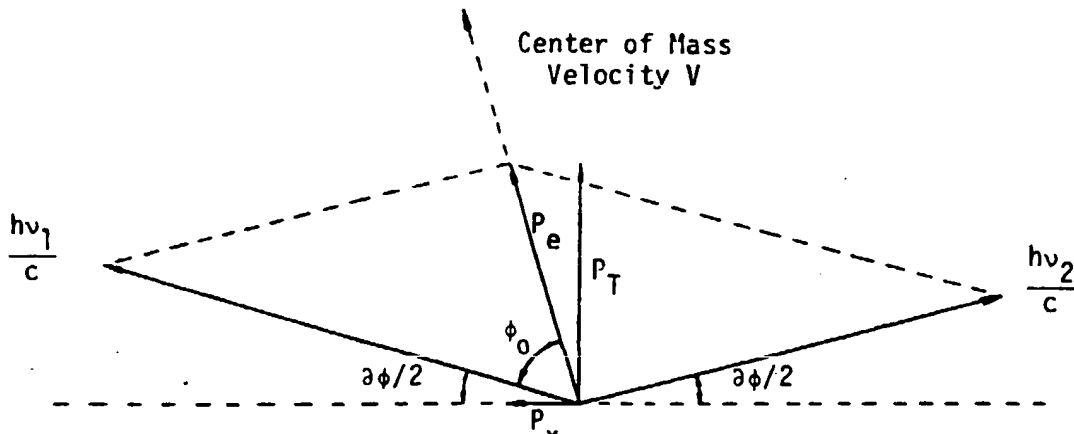
The total stopping time is given by

$$t_s = t_1 + t_2 + t_3 \quad (\text{A.11})$$

with typical theoretical values of 13 psec in Al and 45 psec in Pb (Brandt and Arista 1982).

## APPENDIX B

The purpose of this appendix is to describe the relationships between the momentum distribution of the electrons in the sample and the measured Doppler shifts in energy as well as the angular deviations from colinearity of the  $\gamma$ -rays. In the center of mass rest reference frame the annihilation  $\gamma$ -rays will be emitted in directions separated by  $180^\circ$  and each with an energy of 511 KeV which is equal to the rest mass of an electron (or positron). However, in the laboratory frame the momentum of the electron (the positron is assumed to be at rest as described in the text) produces changes in both the observed  $\gamma$ -ray energy and the angle between the  $\gamma$ -rays. The geometry of the situation is shown in the diagram below.



Straightforward application of the Lorentz transformations of special relativity combined with some trigonometric manipulation yield the result that

$$\delta\phi \approx \left(\frac{2v}{c}\right) \sin \phi_0 + \theta \left(\frac{v^2}{c^2}\right) \quad (\text{B.1})$$

where  $\delta\phi$  is the angular deviation from  $180^\circ$

$v$  is the electron velocity

$c$  is the speed of light

$\phi_0$  is the angle between the electron velocity vector and the direction of  $\gamma$ -ray emission

and  $\theta(\frac{v^2}{c^2})$  is a term of order  $\frac{v^2}{c^2}$  which is negligible for  $v \ll c$

(For a detailed derivation of this result see Rindler 1960). The relativistic definition of momentum is

$$p = \alpha m_0 v \quad (B.2)$$

where  $p$  is the momentum

$$\alpha = (1 - \frac{v^2}{c^2})^{-1/2}$$

and  $m_0$  is the particle mass

For  $v \ll c$ ,  $\alpha \rightarrow 1$  and  $p = mv$

Substituting this result into Eq. (B.1) yields

$$\delta\phi = \frac{2p}{m_0 c} \sin \phi_0 \quad (B.3)$$

Defining the transverse momentum component as

$$p_T = p \sin \phi_0 \quad (B.4)$$

and substituting Eq. (B.4) into Eq. (B.3) yields the final result

$$\delta\phi = \frac{2p_T}{m_0 c} \quad (B.5)$$

demonstrates that measuring the distribution of the angular deviations,  $\delta\phi$ , is in essence equivalent to measuring the distribution of the momentum of the electrons within the sample.

In the case of Doppler measurements one has

$$\delta p_\gamma = p_e \cos \phi_0 \quad (B.6)$$

where  $\delta p_\gamma$  is the change in momentum of the  $\gamma$ -ray

$p_e$  is the electron momentum

and  $\phi_0$  is as defined above

Using Eq. (B.2) with  $v \ll c$  Eq. (B.6) takes the form

$$\delta p_\gamma = m_e v_e \cos \phi_0 \quad (B.7)$$

It can be shown that in relativistic dynamics

$$E^2 = p^2 c^2 + m_0 c^2 \quad (B.8)$$

where  $E$  is the relativistic energy

$p$  is the relativistic momentum

$c$  is the speed of light

and  $m_0$  is the particle rest mass

Now for a photon ( $\gamma$ -ray)  $m_0 = 0$  so Eq. (B.8) reduces to

$$E_\gamma = p_\gamma c \quad (B.9)$$

Combining Eqs. (B.6), (B.7), and (B.9) yields

$$\delta E_\gamma = \delta p_\gamma c = p_e (\cos \phi_0) c = m_e c v_e (\cos \phi_0) \quad (B.10)$$

where  $\delta E_\gamma$  is the Doppler energy shift of the observed  $\gamma$ -ray

$\delta p_\gamma$  is the change in momentum of the  $\gamma$ -ray

and  $p_e$  is the momentum of the annihilating electron

From Eq. (B.10) it can be seen that the momentum distribution of the annihilating electrons can be deduced from the measured Doppler energy shifts.

Combining Eq. (B.5) with (B.10) yields the following relationship between  $\delta\phi$  and  $\delta E$ :

$$\delta E = 255.5 \delta\phi \quad (B.11)$$

where  $\delta E$  is in eV

and  $\delta\phi$  is in mrad

Finally, by combining Eq. (B.10) and the fact that for an electron  $m_0 c^2 = 511$

KeV it can be shown that

$$\delta E_{\gamma} = 511(\cos \phi_0) \left( \frac{v_e}{c} \right) \quad (\text{B.12})$$

where  $\delta E_{\gamma}$  is in KeV

For the case of a 1 eV electron ( $v_e = 5.93 \times 10^7$  cm/sec) moving in the direction of  $\gamma$ -ray emission, Eq. B.12 predicts a 1.01 KeV shift in observed energy.



## APPENDIX C

The material examined in this work was high purity (99.9995%) aluminum supplied by the Materials Research Corporation. The chemical analysis, in terms of PPM of metallic interstitials (and some gases) is listed below.

<u>Element</u>	<u>PPM</u>	<u>Element</u>	<u>PPM</u>	<u>Element</u>	<u>PPM</u>
Ag	<0.1	Li	<0.1	U	
Al	major	Mg	1.5	V	<0.1
As	<0.1	Mn	<0.1	W	<0.2
Au	<0.1	Mo	<0.1	Y	
B		Na	0.2	Zn	0.4
Ba		Nb	<0.1	Zr	<0.1
Be		Ni	0.1		
Bi	<0.1	Pb	<0.1	Cl	0.6
Ca	<0.1	Pd	<0.1	F	
Cd	<0.1	Pt	<0.14	H	3
Co	<0.1	Rh	<0.1	N	5
Cr	<0.1	Sb	<0.1	O	20
Cu	0.1	Si	0.9	P	0.3
Fc	<0.1	Sn	<0.3	S	
Ga	<0.1	Ta			
Ge	<0.1	Th			
In	<0.1	Ti	<0.1		
K	<0.1	Tl			

## APPENDIX D

In this Appendix the utility of Fourier analysis in the solution of the convolution integral equation will be demonstrated. In the first section of Chapter III it was shown that

$$h(y) = \int_{-\infty}^{\infty} f(x)r(y - x)dx \quad (D.1)$$

By definition of the Fourier transform

$$F(w) = \int_{-\infty}^{\infty} e^{iwx}f(x)dx \quad (D.2)$$

Therefore

$$F(w)R(w) = \left[ \int_{-\infty}^{\infty} e^{iwx}f(x)dx \right] \left[ \int_{-\infty}^{\infty} e^{iwt}r(t)dt \right] \quad (D.3)$$

$$= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{iwx}f(x)e^{iwt}r(t)dxdt \quad (D.4)$$

$$= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{iw(x+t)}f(x)r(t)dxdt \quad (D.5)$$

Now let  $x + t = y$

so  $t = y - x$

and  $dt = dy$

Hence Eq. (D.5) can be rewritten as

$$F(w)R(w) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{iwy}f(x)r(y - x)dx dy \quad (D.6)$$

$$= \int_{-\infty}^{\infty} e^{iwy} \left[ \int_{-\infty}^{\infty} f(x)r(y - x)dx \right] dy \quad (D.7)$$

But the integral within the brackets is by Eq. (D.1) equal to  $h(y)$ . Therefore Eq. (D.7) can be rewritten as

$$F(w)R(w) = \int_{-\infty}^{\infty} e^{iwy}h(y)dy \quad (D.8)$$

and using Eq. (D.2) on the right hand side

$$F(w)R(w) = H(w) \quad (D.9)$$

This demonstrates that in Fourier space convolution is simply a complex multiplication.

## APPENDIX E

The purpose of this Appendix is to demonstrate some of the mathematical difficulties associated with the incorrectly posed convolution integral equation. The primary source of the problem is that the left hand side of the equation

$$h^*(x) = \int_a^b f(y)r(x-y)dy \quad (E.1)$$

is only known to within some error  $\epsilon$ . That is, if  $h(x)$  is the "true" function then

$$\max_{a < x < b} |h^*(x) - h(x)| < \epsilon \quad (E.2)$$

Any function  $f_i(y)$  which when convoluted with the known resolution function  $r(x)$  yields a function  $h_i^*(x)$  that satisfies Eq. (E.2) must be considered a solution of Eq. (E.1).

Assume there are two solutions to Eq. (E.1) which are  $f_1(x)$  and  $f_2(x) = f_1(x) + c \sin(2\pi n x / (b-a))$ . By definition

$$h_1^*(x) = \int_a^b f_1(y)r(x-y)dy \quad (E.3)$$

and

$$h_2^*(x) = \int_a^b [f_1(y) + C \sin(2\pi n y / (b-a))]r(x-y)dy \quad (E.4)$$

Subtracting Eq. (E.3) from Eq. (E.4) yields

$$h_2^*(x) - h_1^*(x) = \int_a^b r(x-y) C \sin(2\pi ny/(b-a)) dy \quad (E.5)$$

Now integrating by parts with

$$\begin{aligned} u &= r(x-y) & dv &= C \sin\left(\frac{2\pi Ny}{b-a}\right) dy \\ du &= \frac{d(r(x-y))}{dy} dy & v &= -\frac{C(b-a)}{2\pi N} \cos\left(\frac{2\pi Ny}{b-a}\right) \end{aligned}$$

yields

$$\begin{aligned} \int_a^b r(x-y) C \sin\left(\frac{2\pi Ny}{b-a}\right) dy &= r(x-y) \left[ -\frac{C(b-a)}{2\pi N} \cos\left(\frac{2\pi Ny}{b-a}\right) \right]_a^b \\ &\quad - \int_a^b \frac{-C(b-a)}{2\pi N} \cos\left(\frac{2\pi Ny}{b-a}\right) \frac{d((x-y))}{dy} dy \end{aligned} \quad (E.6)$$

Now integrating the last term on the right hand side by parts with

$$\begin{aligned} u &= \cos\left(\frac{2\pi Ny}{b-a}\right) & dv &= \frac{d(r(x-y))}{dy} dy \\ du &= -\left(\frac{2\pi Ny}{b-a}\right) \sin\left(\frac{2\pi Ny}{b-a}\right) dy & v &= r(x-y) \end{aligned}$$

Yields

$$\begin{aligned} \int_a^b \cos\left(\frac{2\pi Ny}{b-a}\right) \frac{d(r(x-y))}{dy} dy &= [r(x-y) \cos\left(\frac{2\pi Ny}{b-a}\right)]_a^b \\ &\quad - \int_a^b -r(x-y) \left(\frac{2\pi Ny}{b-a}\right) \sin\left(\frac{2\pi Ny}{b-a}\right) dy \end{aligned} \quad (E.7)$$

Combining Eqs. (E.6) and (E.7) yields

$$\int_a^b f(x-y) C \sin\left(\frac{2\pi Ny}{b-a}\right) dx = \frac{-C(b-a)}{2\pi N} [r(x-y) \cos\left(\frac{2\pi Ny}{b-a}\right)]_a^b \quad (E.8)$$

and combining Eqs. (E.8) and (E.5) yields the result

$$h_2^*(x) - h_1^*(x) = \frac{C}{N} \left[ \left( \frac{a-b}{2\pi} \right) r(x-y) \cos\left(\frac{2\pi Ny}{b-a}\right) \right]_a^b \quad (E.9)$$

So it can be seen that for any value of C one can select a value of N large

enough so that the difference between  $h_2^*(x)$  and  $h_1^*(x)$  is smaller than any previously determined number. Therefore, both  $h_1^*(x)$  and  $h_2^*(x)$  satisfy Eq. (E.2) and  $f_1(x)$  as well as  $f_2(x)$  must be solutions to Eq. (E.1). The important point is that one may select  $C$  and  $N$  in such a way as to allow  $f_1(x)$  and  $f_2(x)$  to differ from each other by as much as one likes. Clearly the problem as posed does not have a unique solution and a different solution technique is required.

# APPENDIX F

```

CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC
C
C   THIS IS A LISTING OF THE FORTRAN 77 VERSION OF THE PROGRAM C
C   DECONFFT WHICH IS DESIGNED TO DECONVOLUTE DOPPLER BROADENED C
C   POSITRON ANNIHILATION SPECTRA. FOR A DESCRIPTION OF THE C
C   MATHEMATICS INVOLVED SEE J.P. SCHAPPER, E.J. SHAUGHNESSY, C
C   AND P.L. JONES, JOURNAL OF NUCLEAR INSTRUMENTS AND METHODS C
C   IN PHYSICS RESEARCH, B5, 74, 1984. C
C
CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC

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      INTEGER PCSP
      CHARACTER*2 PCSZ,PCH
      CHARACTER*80 XCH,YCH,TCH
      CHARACTER*7 NAME,NAMEE
      DIMENSION IA(2048), IB(2048), DOPE(12),XP(74),YP(74)
      DOUBLE PRECISION A(2048),B(2048),C(2048),FR(2048),HC(2048)
      DOUBLE PRECISION D(2048),E(2048),G(2),S(2048),SI(2048)
      DOUBLE PRECISION F1(2048),PI,V,W,WI
      COMPLEX*16 R(4096),H(4096),F(4096),T,A1,H1(4096)
      COMPLEX*16 A2,R1(4096),T1
      PI=3.141592653589793D0
      PRINT*,'THIS PROGRAM DEALS WITH THE CONVOLUTION'
      PRINT*,'H(X)=F(X)*R(X), WHERE R IS THE'
      PRINT*,'RESOLUTION FUNCTION, H IS THE'
      PRINT*,'BROADENED AND OBSERVED FUNCTION, AND'
      PRINT*,'F IS THE TRUE AND DESIRED FUNCTION.'
      PRINT*,' '
C   THE RESOLUTION FUNCTION IS USUALLY OBTAINED FROM
C   THE Sr(85) GAMMA RAY (514 KeV).
C
      2 PRINT*,'THE PROGRAM REQUIRES N DATA POINTS'
      PRINT*,'WHERE N=2**M, AND M IS AN INTEGER.'
      PRINT*,' '
C   THESE DATA POINTS WILL BE H(X) AND R(X) AS COLLECTED
C   BY THE MCA.
C
      PRINT*,'ENTER THE VALUE OF M TO BE USED IN THIS'
      PRINT*,'PROGRAM. (M=11 YIELDS N=2048)'
      PRINT*,' '
      READ(*, '(I2)') M
      N=2**M
      ND2=N/2
      ND4=N/4
      PRINT*,'ENTER THE RESOLUTION FUNCTION FILENAME'
      PRINT*,' '
      READ (*, '(A7)') NAME
      OPEN (90,FILE=NAME,PAD='YES')
      READ (90,1) (IA(I), I=1,2048)
1  FORMAT=(5X,5I7)
      CLOSE (90)

```

```

PRINT*, 'ENTER THE EXPERIMENTAL FUNCTION FILENAME'
PRINT*, '
READ (*, '(A7)') NAME
OPEN (90, FILE=NAME, PAD='YES')
READ (90, 1) (IB(I), I=1, 2048)
CLOSE (90)
NIN=2048
C THE SUBROUTINE DELSKIP DELETES THE LINE
C SKIP IN THE RAW DATA SUPPLIED BY THE MCA.
  CALL DELSKIP (IA, A, NIN)
  CALL DELSKIP (IB, B, NIN)
C THE FOLLOWING SUBROUTINE CALCULATES THE CENTRAL
C CHANNEL LOCATION BASED ON THE CENTER OF THE
C 11 CHANNEL REGION WHICH CONTAINS THE MAXIMUM
C TOTAL NUMBER OF COUNTS
  CALL CENTER (A, IMAXA)
  CALL CENTER (B, IMAXB)
C
C THIS NEXT LARGE SECTION DEALS WITH THE
C BACKGROUND CORRECTION.
C
  PRINT*, 'DO YOU WISH TO BACKGROUND CORRECT? '
  READ (*, '(A1)') NAME
  IF (NAME .EQ. 'Y') GO TO 3
  CALL NOBACK (N, B, IMAXB)
  GO TO 200
3 PRINT*, 'DO YOU WISH TO BACKGROUND CORRECT USING'
  PRINT*, 'THE JACKMAN METHOD OR THE SPECTRAL FILTERING'
  PRINT*, 'TECHNIQUE? (J OR S) '
  READ (*, '(A1)') NAME
  IF (NAME .EQ. 'J') GO TO 5
  PRINT*, 'THE NEXT SEVERAL QUESTIONS REFER TO THE'
  PRINT*, 'RESOLUTION FUNCTION. '
  CALL BACK (A, IMAXA, N)
  PRINT*, 'THE NEXT SEVERAL QUESTIONS REFER TO THE'
  PRINT*, 'EXPERIMENTAL FUNCTION. '
  CALL BACK (B, IMAXB, N)
  GO TO 377
5 PRINT*, 'ENTER THE RESOLUTION OF THE MCA IN'
  PRINT*, 'eV/CHANNEL. (USE A DECIMAL POINT)'
  PRINT*, '(SUGGESTION: IF THE SYSTEM IS SET'
  PRINT*, 'IN ITS NORMAL MODE USE 64. )'
  PRINT*, '
  READ (*, '(P6.2)') EV
  X=(1000.D0/EV)+0.5D0
  IK=IFIX (X)
  IKLA=(IMAXA-20)/IK
  ILA=514-IKLA
  IKHA=(1024-IMAXA)/IK
  IHA=514+IKHA
  IKLB=(IMAXB-20)/IK

```



```

ILB=511-IKLB
IKHB=(1024-IMAXB)/IK
IHB=511+IKHB
PRINT*, 'THE EXPERIMENTAL SPECTRUM HAS BEEN COLLECTED'
PRINT*, 'OVER THE FOLLOWING KeV REGION:'
WRITE (*,10) ILB,IHB
10 FORMAT (I5,I5)
PRINT*, 'WE ARE NOW READY TO PROCEED WITH THE'
PRINT*, 'BACKGROUND CORRECTION. OVER WHICH'
PRINT*, 'LOW ENERGY REGION WOULD YOU LIKE THE'
PRINT*, 'LINEAR LEAST SQUARE FIT? IF POSSIBLE'
PRINT*, 'SELECT 486 TO 497. THE PROPER FORM'
PRINT*, 'OF ENTRY IS TWO 3 DIGIT INTEGERS'
PRINT*, 'SEPARATED BY TWO BLANKS.'
PRINT*, ' '
READ (*,*) JLAB,JLBB
IS=IMAXB-(511-JLAB)*IK
IF=IMAXB-(511-JLBB)*IK
N1=IF-IS
DO 20 I=1,N1
    J=IS-1+I
    D(I)=J
    E(I)=B(J)
20 CONTINUE
CALL FIT (N1,D,E,G)
BBL=G(1)
XMBL=G(2)
PRINT*, 'OVER WHICH HIGH ENERGY REGION WOULD'
PRINT*, 'YOU LIKE THE LEAST SQUARE FIT?'
PRINT*, 'IF POSSIBLE SELECT 534 542.'
PRINT*, ' '
READ (*,*) JHAB,JHBB
IS=IMAXB+(JHAB-511)*IK
IF=IMAXB+(JHBB-511)*IK
N1=IF-IS
DO 30 I=1,N1
    J=IS-1+I
    D(I)=J
    E(I)=B(J)
30 CONTINUE
CALL FIT (N1,D,E,G)
BBH=G(1)
XMBH=G(2)
PRINT*, 'TO WHICH ENERGIES WOULD YOU LIKE TO'
PRINT*, 'EXTRAPOLATE THE LINEAR BACKGROUND'
PRINT*, 'CORRECTION? (SUGGESTION: 503 518)'
PRINT*, ' '
READ (*,*) IL,IH
PRINT*, 'ON THE HIGH ENERGY SIDE OF THE PEAK,'
PRINT*, 'WHAT IS THE HIGHEST ENERGY YOU WISH'
PRINT*, 'TO HAVE CONSIDERED FOR DECONVOLUTION?'

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PRINT*, 'SUGGESTION: 540'
PRINT*, '
READ (*,*) IEND
IL=IMAXB-(511-IL)*IK
IH=IMAXB+(IH-511)*IK
IEND=IMAXB+(IEND-511)*IK
IEB=IEND
DO 35 I=IH, IEND
    B(I)=B(I)-(XMBH*I+BBH)
    IF (B(I) .LT. 0.) B(I)=0.D0
35 CONTINUE
    XH=XMBH*IH+BBH
    XL=XMBL*IL+BBL
    XM=(XL-XH)/(FLOAT(IL-IH))
    XB=XH-((XL-XH)/(FLOAT(IL-IH)))*IH
    DO 40 I=IMAXB, IH
        B(I)=B(I)-(XM*I+XB)
        IF (B(I) .LT. 0.) B(I)=0.D0
40 CONTINUE
    PRINT*, 'THE RESOLUTION SPECTRUM HAS BEEN COLLECTED'
    PRINT*, 'OVER THE FOLLOWING KeV REGION:'
    WRITE (*,10) ILA, IHA
    PRINT*, 'OVER WHICH LOW ENERGY REGION WOULD YOU LIKE'
    PRINT*, 'THE LINEAR LEAST SQUARE FIT?'
    PRINT*, 'IF POSSIBLE SELECT 489 500.'
    PRINT*, '
    READ (*,*) JLAA, JLBA
    IS=IMAXA-(514-JLAA)*IK
    IF=IMAXA-(514-JLBA)*IK
    N1=IF-IS
    DO 45 I=1, N1
        J=IS-1+I
        D(I)=J
        E(I)=A(J)
45 CONTINUE
    CALL FIT (N1, D, E, G)
    BAL=G(1)
    XMAL=G(2)
    PRINT*, 'OVER WHICH HIGH ENERGY REGION WOULD YOU LIKE'
    PRINT*, 'THE LINEAR LEAST SQUARE FIT?'
    PRINT*, 'IF POSSIBLE SELECT 537 545.'
    PRINT*, '
    READ (*,*) JHAA, JHBA
    IS=IMAXA+(JHAA-514)*IK
    IF=IMAXA+(JHBA-514)*IK
    N1=IF-IS
    DO 50 I=1, N1
        J=IS-1+I
        D(I)=J
        E(I)=A(J)
50 CONTINUE

```

```

CALL FIT (N1,D,E,G)
BAH=G(1)
XMAH=G(2)
PRINT*, 'TO WHICH ENERGIES WOULD YOU LIKE TO'
PRINT*, 'EXTRAPOLATE THE LINEAR BACKGROUND'
PRINT*, 'CORRECTION? (SUGGESTION: 506 521)'
PRINT*, ' '
READ (*,*) IL,IH
PRINT*, 'ON THE HIGH ENERGY SIDE OF THE PEAK,'
PRINT*, 'WHAT IS THE HIGHEST ENERGY YOU WISH'
PRINT*, 'TO HAVE CONSIDERED FOR DECONVOLUTION?'
PRINT*, 'SUGGESTION: 521'
PRINT*, ' '
READ (*,*) IEND
IL=IMAXA-(514-IL)*IK
IH=IMAXA+(IH-514)*IK
IEND=IMAXA+(IEND-514)*IK
IEA=IEND
DO 55 I=IH,IEND
  A(I)=A(I)-(XMAH*I+BBH)
  IF (A(I) .LT. 0.) A(I)=0.D0
55 CONTINUE
  XH=XMAH*IH+BAH
  XL=XMAL*IL+BAL
  XM=(XL-XH)/(FLOAT(IL-IH))
  XB=XH-((XL-XH)/(FLOAT(IL-IH)))*IH
DO 60 I=IMAXA,IH
  A(I)=A(I)-(XM*I+XB)
  IF (A(I) .LT. 0.) A(I)=0.D0
60 CONTINUE
GO TO 378
377 CONTINUE
  IEA=IMAXA+109
  IEB=IMAXB+453
378 CONTINUE
C
C THIS IS THE END OF THE BACKGROUND CORRECTION SEQUENCE.
C NEXT WE NEED TO REFLECT THE HIGH ENERGY HALF OF THE
C BACKGROUND CORRECTED PEAK ABOUT THE CENTRAL POINT
C TO FORM A SYMMETRIC PEAK. IN ADDITION A ZEROFILL
C TO 2048 DATA POINTS IS REQUIRED
C
CALL ZEROFILL (A,IMAXA,IEA,N)
CALL ZEROFILL (B,IMAXB,IEB,N)
M1=IEA-IMAXA+10
CALL TAPER (A,M1,N)
M2=IEB-IMAXB+20
CALL TAPER (B,M2,N)
M3=M1
CALL SMOOTH (A,M3,N)
M4=M2

```

```

      CALL SMOOTH (B,M4,N)
C   THE NEXT TWO DO LOOPS NORMALIZE THE AREA
C   OF THE RESOLUTION AND BROADENED SPECTRA.
      V=0.D0
      W=0.D0
      DO 62 I=1,N
        V=V+A(I)
        W=W+B(I)
62   CONTINUE
      DO 63 I=1,N
        A(I)=A(I)/V
        B(I)=B(I)/W
63   CONTINUE
      DO 68 I=1,N
        S(I)=B(I)
68   CONTINUE
      DO 72 I=1,ND2
        J=ND2+I
        V=S(I)
        S(I)=S(J)
        S(J)=V
72   CONTINUE
      PRINT*, 'DO YOU WISH TO SAVE THE BACKGROUND'
      PRINT*, 'CORRECTED SPECTRUM? '
      READ (*, '(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 76
      PRINT*, 'ENTER THE FILENAME. '
      READ (*, '(A7)') NAME
      OPEN (90, FILE=NAME, PAD='YES')
      WRITE (90,74) (S(I), I=1,N)
74   FORMAT (5X,5D22.10)
      CLOSE (90)
      CALL PARAMS (N,S)
76   CONTINUE
      DO 77 I=1,N
        R(I)=DCMPLX(A(I),0.D0)
        B(I)=DCMPLX(B(I),0.D0)
77   CONTINUE
      CALL REORDER (M,H)
      CALL FFT (M,H)
      CALL REORDER (M,R)
      CALL FFT (M,R)
      CALL DECON (M,P,R,H)
C
      DOPE(1)=150.
      DOPE(2)=150.
      DOPE(3)=950.
      DOPE(4)=650.
      DOPE(5)=0.
      DOPE(6)=0.
      PCSZ='NS'

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```

      PCSP=1
C
C THIS NEXT SMALL SECTION ALLOWS ONE TO VIEW THE POWER
C SPECTRUM IF SO DESIRED.
      PRINT*, 'WOULD YOU LIKE TO SEE AND PLOT THE POWER SPECTRUM'
      PRINT*, 'OF THE EXPERIMENTAL DATA AND THE DECONVOLUTED DATA.'
      PRINT*, ' '
      READ (*, '(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 179
      DO 78 I=1,ND2
        D(I)=H(I)*(CONJG(H(I)))
78 CONTINUE
      PRINT*, 'ENTER THE FILENAME FOR THE P.S. OF THE EX. DATA. '
      PRINT*, ' '
      READ (*, '(A7)') NAME
      OPEN (90, FILE=NAME, PAD='YES')
      WRITE (90,901) (I,D(I),I=1,ND2)
901 FORMAT (5(I4,D22.12))
      CLOSE (90)
      CALL REORDER (M,F)
      CALL FFT (M,F)
      T=0.D0
      DO 277 I=1,N
        T=T+F(I)
277 CONTINUE
      DO 278 I=1,N
        F(I)=F(I)/T
278 CONTINUE
      CALL REORDER (M,F)
      CALL FFT (M,F)
      DO 178 I=1,ND2
        E(I)=F(I)*(CONJG(F(I)))
178 CONTINUE
      PRINT*, 'ENTER THE FILENAME FOR THE P.S. OF THE DECON. DATA.'
      PRINT*, ' '
      READ (*, '(A7)') NAME
      OPEN (90, FILE=NAME, PAD='YES')
      WRITE (90,901) (I,E(I),I=1,ND2)
      CLOSE (90)
      NPTS=74
      DO 910 I=1,10
        XP(I)=ALOG10(FLOAT(I))
        YP(I)=E(I)
        YP(I)=ALOG10(YP(I))
910 CONTINUE
      DO 912 I=15,100,5
        J=8+(I/5)
        XP(J)=ALOG10(FLOAT(I))
        YP(J)=E(I)
        YP(J)=ALOG10(YP(J))
912 CONTINUE

```

```

DO 914 I=120,1000,20
  J=23+(I/20)
  XP(J)=ALOG10(FLOAT(I))
  YP(J)=E(I)
  YP(J)=ALOG10(YP(J))
914 CONTINUE
  XP(74)=ALOG10(1020.)
  YP(74)=-10.
  XCH='LOG OF THE FOURIER COEFFICIENT NO.'
  MX=34
  YCH='LOG OF THE POWER'
  MY=16
  PRINT*,'ENTER THE TITLE OF THIS PLOT. '
  PRINT*,' '
  READ (*,'(A80)') TCH
  PRINT*,'ENTER THE NO. OF CHARACTERS IN THE TITLE. '
  READ (*,'(I2)') NT
  PCH='1 '
  CALL CKSTUP ('TK15',9600)
  CALL CKER
  CALL CKGRAP (DOPE,NPTS,XP,YP,XCH,MX,YCH,MY,TCH,NT,PCSZ
C,PCSP,PCH)
  DO 930 I=1,10
    YP(I)=D(I)
    YP(I)=ALOG10(YP(I))
930 CONTINUE
  DO 932 I=15,100,5
    J=8+(I/5)
    YP(J)=D(I)
    YP(J)=ALOG10(YP(J))
932 CONTINUE
  DO 934 I=120,1020,20
    J=23+(I/20)
    YP(J)=D(I)
    YP(J)=ALOG10(YP(J))
934 CONTINUE
  PCH='3 '
  CALL CKPLT (DOPE,NPTS,XP,YP,PCSZ,PCSP,PCH)
  CALL CKMA (0.,0.,1)
  READ (*,*)
179 CONTINUE
C
C AT THIS STAGE THE RESOLUTION AND EXPERIMENTAL
C SPECTRA HAVE BEEN TRANSFORMED INTO FOURIER SPACE.
C THIS NEXT SMALL SECTION DETERMINES THE NUMBER OF
C POSITIVE DECREASING FOURIER COEFFICIENTS IN THE
C DECONVOLUTED RESULT. THIS INFORMATION CAN THEN
C BE USED IN THE FILTERING ROUTINES IF SO DESIRED.
C
DO 80 I=2,N
  J=I-1

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      D(I)=REAL(P(I))
      D(J)=REAL(P(J))
      IF (D(I) .LT. 0.0) GO TO 81
      IF (D(I) .GT. D(J)) GO TO 81
80 CONTINUE
81 IPD=I-1
      WRITE (*,82) IPD
82 FORMAT (5X,'THE NO. OF POSITIVE DECREASING COEFS. IS ' I5)
      PRINT*, '
      PRINT*, 'DO YOU WISH TO USE A COSINE TAPER FILTER'
      PRINT*, 'TO MODIFY THE FOURIER COEFFICIENTS?'
      READ (*, '(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 189
      PRINT*, 'ENTER THE NUMBER OF THE FIRST COEFFICIENT'
      PRINT*, 'TO BE SET EQUAL TO ZERO. '
      READ (*, '(I3)') IZ
      PRINT*, 'ENTER THE NUMBER OF COEFFICIENTS TO BE TAPERED. '
      READ (*, '(I3)') IT
      IS=IZ-IT
      XS=REAL(IS)
      XZ=REAL(IZ)
      XY=1.0/(XS-XZ)
      DO 181 I=IS, IZ
          X=REAL(I)
          P(I)=P(I)*0.5*(1-COS(PI*XY*(X-XZ)))
181 CONTINUE
      J=IZ+1
      K=(N/2)+1
      DO 183 I=J, K
          P(I)=DCMLPX (0.D0,0.D0)
183 CONTINUE
      K=K-1
      DO 185 I=2, K
          J=N+2-I
          F(J)=F(I)
185 CONTINUE
189 PRINT*, 'DO YOU WISH TO USE A BOXCAR FILTER TO'
      PRINT*, 'MODIFY THE FOURIER COEFFICIENTS IN'
      PRINT*, 'ORDER TO REMOVE THE HIGH FREQ. NOISE?'
      READ (*, '(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 84
      PRINT*, 'ENTER THE NUMBER OF COEFFICIENTS TO BE USED: '
      READ (*, *) J
      J=J+1
      K=N+2-J
      DO 83 I=J, K
          F(I)=DCMLPX(0.D0,0.D0)
83 CONTINUE
84 CONTINUE
      PRINT*, 'DO YOU WANT TO SEE THE FOURIER COEFFICIENTS?'
      READ (*, '(A1)') NAME

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```

      IF (NAME .EQ. 'N') GO TO 85
      PRINT*, 'ENTER THE FILENAME FOR H(X). '
      PRINT*, ' '
      READ (*, '(A7)') NAME
      OPEN (90, FILE=NAME, PAD='YES')
      WRITE (90, 911) (I, H(I), I=1, N)
911  FORMAT (2(I4, 2D22.10))
      CLOSE (90)
      OPEN (90, FILE='PCSF', PAD='YES')
      WRITE (90, 911) (I, F(I), I=1, N)
      CLOSE (90)
85  CONTINUE
      CALL REORDER (M, F)
      CALL FFT (M, F)
      CALL REORDER (M, H)
      CALL FFT (M, H)
C   THIS NEXT SECTION ALLOWS ONE TO USE THE ITERATION
C   ROUTINE TO IMPROVE F(X).
      PRINT*, 'WOULD YOU LIKE TO USE THE ITERATION'
      PRINT*, 'SUBROUTINE TO TRY TO IMPROVE THE'
      PRINT*, 'ACCURACY OF THE DECONVOLUTED SPECTRUM? '
      READ (*, '(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 86
      CALL ITER (M, R, F, H)
86  CONTINUE
C   THIS NEXT DO LOOP SHIFTS THE PHASE OF THE
C   EXPERIMENTAL FUNCTION H, AND THE DECONVOLUED
C   FUNCTION F, SO THAT THEIR MAXIMA ARE LOCATED
C   IN THE CENTRAL CHANNELS.
      ND2=N/2
      DO 88 I=1, ND2
          J=ND2+I
          T=F(I)
          T1=H(I)
          F(I)=F(J)
          H(I)=H(J)
          F(J)=T
          H(J)=T1
88  CONTINUE
      DO 98 I=1, N
          F1(I)=REAL(F(I))
98  CONTINUE
      W=0.D0
      DO 99 I=1, N
          W=W+F1(I)
99  CONTINUE
      DO 100 I=1, N
          F1(I)=F1(I)/W
100 CONTINUE
      PRINT*, 'DO YOU WANT A PLOT OF h(X) AND f(X)? '
      READ (*, '(A1)') NAME

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```

      IF (NAME .EQ. 'N') GO TO 105
      NPTS=60
      W=F1(1025)
      DO 102 I=1,60
        J=1024+I
        XP(I)=.064*(FLOAT(I))
        YP(I)=F1(J)/W
102  CONTINUE
      XCH='KeV'
      MX=3
      YCH='h(X) AND f(X)'
      MY=13
      PRINT*, 'ENTER THE TITLE OF THIS PLOT.'
      PRINT*, ' '
      READ (*, '(A80)') TCH
      PRINT*, 'ENTER THE NO. OF CHARACTERS IN THE TITLE. '
      READ (*, '(I2)') NT
      PCH='1 '
      CALL CKSTUP ('TK15',9600)
      CALL CKER
      CALL CKGRAP (DOPE,NPTS,XP,YP,XCH,MX,YCH,MY,TCH,NT,PCSZ
C,PCSP,PCH)
      DO 103 I=1,60
        J=1024+I
        YP(I)=S(J)/W
103  CONTINUE
      PCH='3 '
      CALL CKPLT (DOPE,NPTS,XP,YP,PCSZ,PCSP,PCH)
      CALL CKMA (0.,0.,1)
      READ (*,*)
105  PRINT*, 'DO YOU WISH TO SAVE THE '
      PRINT*, 'DECONVOLUTED SPECTRUM? '
      READ (*, '(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 101
      PRINT*, 'ENTER THE FILENAME. '
      READ (*, '(A7)') NAME
      OPEN (90, FILE=NAME, PAD='YES')
      WRITE (90,74) (F1(I), I=1,N)
      CLOSE (90)
101  CONTINUE
      PRINT*, 'DO YOU WISH TO CHECK YOUR RESULT BY'
      PRINT*, 'CONVOLUTING f(X) WITH r(X) AND COMPARING'
      PRINT*, 'THE RESULT WITH h(X)?'
      READ (*, '(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 200
      CALL REORDER (M,F)
      CALL FFT (M,F)
      CALL CONV (M,F,R,H1)
      CALL REORDER (M,H1)
      CALL FFT (M,H1)
      DO 106 I=1,N

```

```

      HC(I)=REAL(H1(I))
106 CONTINUE
      V=0.D0
      DO 107 I=1,N
        V=V+HC(I)
107 CONTINUE
      DO 108 I=1,N
        HC(I)=HC(I)/V
108 CONTINUE
      PRINT*,'ENTER THE FILENAME FOR THE RESULT'
      PRINT*,'OF THIS COMPARISON.'
      PRINT*,' '
      READ (*,'(A7)') NAME
      OPEN (90,FILE=NAME,PAD='YES')
      WRITE (90,110)
110 FORMAT (3X,'I',14X,'h(X) RAW',14X,'h(X)=(F*R)',14X,'f(X)')
      WRITE (90,120) (I,S(I),HC(I),F1(I),I=1025,1300)
120 FORMAT (1X,I4,2X,3D22.10)
      CLOSE (90)
      PRINT*,'DO YOU WANT A PLOT OF h(X) AND THE ESTIMATE'
      PRINT*,'OF h(X) OBTAINED FROM f(X)*r(X)? '
      READ (*,'(A1)') NAME
      IF (NAME .EQ. 'N') GO TO 200
      NPTS=60
      W=HC(1025)
      DO 130 I=1,60
        J=1024+I
        XP(I)=.064*(FLOAT(I))
        YP(I)=HC(J)/W
130 CONTINUE
      XCH='KeV'
      MX=3
      YCH='h(X) AND h` (X)'
      MY=14
      PRINT*,'ENTER THE TITLE OF THIS PLOT.'
      PRINT*,' '
      READ (*,'(A80)') TCH
      PRINT*,'ENTER THE NO. OF CHARACTERS IN THE TITLE. '
      READ (*,'(I2)') NT
      PCB='1 '
      CALL CKSTUP ('TK15',9600)
      CALL CKER
      CALL CKGRAF (DOPE,NPTS,XP,YP,XCH,MX,YCH,MY,TCH,NT,PCSZ
C,PCSP,PCB)
      DO 140 I=1,60
        J=1024+I
        YP(I)=S(J)/W
140 CONTINUE
      PCH='3 '
      CALL CKPLT (DOPE,NPTS,XP,YP,PCSZ,PCSP,PCH)
      CALL CKMA (0.,0.,1)

```

```

      READ (*,*)
200  CONTINUE
      CALL PARAMS (N,F1)
      STOP
      END
CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC
C
      SUBROUTINE DELSKIP (IC,C,NIN)
      DIMENSION IC(2048)
      DOUBLE PRECISION C(1024)
      J=0
      K=0
      DO 10 I=1,NIN
        J=J+1
        K=K+1
        IC(I)=IC(K)
        IF (J .EQ. 5) THEN
          J=0
          K=K+5
        ENDIF
10    CONTINUE
      NID2=NIN/2
      DO 20 I=1,NID2
        C(I)=FLOAT(IC(I))
20    CONTINUE
      RETURN
      END
C
      SUBROUTINE CENTER (A,IMAX)
      DOUBLE PRECISION A(1024),X,T(21),Y
      X=0.D0
      DO 10 I=300,800
        IF (A(I) .GT. X) THEN
          X=A(I)
          J=I-10
        ENDIF
10    CONTINUE
      K=J+21
      Y=0.D0
      DO 20 I=J,K
        L=I-5
        M=I+5
        T(I)=0.D0
        DO 15 N=L,M
          T(I)=T(I)+A(N)
15    CONTINUE
        IF (T(I) .GT. Y) THEN
          Y=T(I)
          IMAX=I
        ENDIF
20    CONTINUE

```

RETURN  
END

C

```

      SUBROUTINE FIT (N,X,Y,C)
C   THIS SUBROUTINE IS A SIMPLIFICATION OF
C   A LARGER SUBROUTINE WHICH FITS A CURVE USING
C   UP TO A NINTH ORDER POLYNOMIAL.  FOR A
C   REFERENCE SEE JPS DISSERTATION.
      DOUBLE PRECISION X(300),Y(300),C(2),A(3,3),XN(300)
      DO 10 I=1,N
10    XN(I)=1.D0
      DO 30 I=1,2
          A(I,1)=0.D0
          A(I,3)=0.D0
          DO 20 J=1,N
              A(I,1)=A(I,1)+XN(J)
              A(I,3)=A(I,3)+Y(J)*XN(J)
20    XN(J)=XN(J)*X(J)
30    CONTINUE
      A(2,2)=0.D0
      DO 40 J=1,N
          A(2,2)=A(2,2)+XN(J)
40    XN(J)=XN(J)*X(J)
      A(1,2)=A(2,1)
      DO 50 I=1,2
          SUM=0.D0
          IF (2 .GT. I) GO TO 45
          SUM=SUM+A(I,1)*A(1,2)
          A(I,2)=A(I,2)-SUM
          GO TO 50
45    IF (ABS(A(I,I)) .LT. 1.E-10) GO TO 99
          A(I,2)=(A(I,2)-SUM)/A(I,I)
50    CONTINUE
      DO 90 J=1,2
90    C(J)=A(J,3)
          C(1)=C(1)/A(1,1)
          SUM=A(2,1)*C(1)
          C(2)=(C(2)-SUM)/A(2,2)
          SUM=A(1,2)*C(2)
          C(1)=C(1)-SUM
      RETURN
99    PRINT*, 'TROUBLE IN SUBROUTINE FIT'
      RETURN
      END

```

C

```

      SUBROUTINE ZEROFILL (C,IM,IE,N)
      DOUBLE PRECISION C(2048),D(2048),T
      M=IE-IM
      MP1=M+1
      DO 10 I=1,N
          D(I)=0.D0

```

```

10 CONTINUE
   DO 20 I=1,MP1
      J=IM+I-1
      D(I)=C(J)
20 CONTINUE
   DO 30 I=1,N
      C(I)=D(I)
30 CONTINUE
   RETURN
   END

```

C

```

SUBROUTINE TAPER (C,M,N)
DOUBLE PRECISION C(2048),T
T=1.D0/1000.D0
DO 90 I=1,M
   IF (C(I) .NE. 0.D0) GO TO 80
   J=I-1
   K=I+1
   C(I)=(C(J)+C(K))/2.D0
   IF (C(I) .LT. T) GO TO 95
80 CONTINUE
90 CONTINUE
   GO TO 97
95 IK=I
   DO 96 J=IK,M
      K=J-1
      C(J)=0.95D0*C(K)
96 CONTINUE
97 CONTINUE
   ND2=N/2.D0
   DO 100 I=2,ND2
      J=ND2+I
      K=ND2+2-I
      C(J)=C(K)
100 CONTINUE
   RETURN
   END

```

C

```

SUBROUTINE SMOOTH (C,M,N)
DOUBLE PRECISION C(2048)
DO 20 I=2,M
   J=I-1
   IF (C(J) .GT. C(I)) GO TO 10
   K=I+1
   C(I)=(C(J)+C(K))/2.D0
5   IF (C(J) .GT. C(I)) GO TO 10
   C(I)=0.999*C(I)
   GO TO 5
10 CONTINUE
20 CONTINUE
   ND2=N/2

```

```

      DO 30 I=2,ND2
        J=ND2+I
        K=ND2+2-I
        C(J)=C(K)
30  CONTINUE
    RETURN
  END

C
  SUBROUTINE REORDER (M,C)
C  IN ORDER TO USE THE FOLLOWING FFT ONE MUST
C  FIRST REORDER THE DATA.  SEE JPS DISSERTATION
C  FOR DETAILS.
    COMPLEX*16 C(4096),T
    N=2**M
    MV2=N/2
    NM1=N-1
    J=1
    DO 7 I=1,NM1
      IF (I .GE. J) GO TO 5
      T=C(J)
      C(J)=C(I)
      C(I)=T
5     K=MV2
6     IF (K .GE. J) GO TO 7
      J=J-K
      K=K/2
      GO TO 6
7    J=J+K
    RETURN
  END

C
  SUBROUTINE FFT (M,A)
C  FOR DETAILS SEE JPS DISSERTATION
    DOUBLE PRECISION PI
    COMPLEX*16 A(4096),U,W,T
    PI=3.141592653589793D0
    N=2**M
    DO 30 L=1,M
      LE=2**L
      LEI=LE/2
      U=(1.D0,0.D0)
      W=DCMPLX(DCOS(PI/LEI),-DSIN(PI/LEI))
      DO 20 J=1,LEI
        DO 10 I=J,N,LE
          IP=I+LEI
          T=A(IP)*U
          A(IP)=A(I)-T
          A(I)=A(I)+T
10     CONTINUE
        U=U*W

```

```

20   CONTINUE
30   CONTINUE
    RETURN
    END

```

C

```

    SUBROUTINE CONV (M,F,R,H)
    DOUBLE PRECISION X
    COMPLEX*16 H(4096),F(4096),R(4096)
    N=2**M
    W1=FLOAT(N)
    DO 10 I=1,N
        H(I)=P(I)*R(I)
10   CONTINUE
    DO 20 I=1,N
        H(I)=H(I)/W1
20   CONTINUE
    RETURN
    END

```

C

```

    SUBROUTINE DECON (M,F,R,H)
    DOUBLE PRECISION X
    COMPLEX*16 F(4096),R(4096),H(4096)
    N=2**M
C   IN FOURIER SPACE DECONVOLUTION IS SIMPLY A
C   COMPLEX DIVISION.
    DO 10 I=1,N
        F(I)=H(I)/R(I)
10   CONTINUE
    X=FLOAT(N)
    DO 30 I=1,N
        F(I)=F(I)/X
30   CONTINUE
    RETURN
    END

```

C

```

    SUBROUTINE ITER (M,R,F,H)
    DIMENSION A(2048),B(2048)
    COMPLEX*16 R(4096),F(4096),H(4096),H1(4096),T
    CHARACTER*7 NAME
    PRINT*,'ENTER THE VALUE FOR THE PARAMETER WHICH'
    PRINT*,'CONTROLS THE ALLOWABLE ERROR BETWEEN THE'
    PRINT*,'OBSERVED SPECTRUM AND F*R. '
    PRINT*,'EPS=THE SORT OF THE SUM OF THE SQRS OF'
    PRINT*,'THE DIFFERENCES BETWEEN THE H(I)S AND'
    PRINT*,'THE F(I)*R(I)S. '
    PRINT*,'(A REAL NUMBER PLEASE.) '
    READ (*,'(F13.7)') EPS
    PRINT*,'ENTER THE MAXIMUM NUMBER OF ITERATIONS'
    PRINT*,'YOU WISH TO PERMIT. '
    PRINT*,'(AN INTEGER PLEASE) '
    READ (*,'(I3)') IEND

```

```

N=2**M
ND2=N/2
ND4=N/4
DO 30 I=1,IEND
  CALL REORDER (M,P)
  CALL PFT (M,P)
  CALL CONV (M,P,R,H1)
  CALL REORDER (M,P)
  CALL FFT (M,P)
  CALL REORDER (M,H1)
  CALL PFT (M,H1)
  RSQ=0.0
  DO 2 IA=1,N
    A(IA)=REAL(H(IA))
    B(IA)=REAL(H1(IA))
2  CONTINUE
  V=0.0
  W=0.0
  DO 4 IAA=1,N
    V=V+A(IAA)
    W=W+B(IAA)
4  CONTINUE
  DO 6 IAB=1,N
    A(IAB)=A(IAB)/V
    B(IAB)=B(IAB)/W
6  CONTINUE
  DO 10 IAC=1,ND4
    RSQ=RSQ+(A(IAC)-B(IAC))**2
10 CONTINUE
  RSQ=RSQ**0.5
  WRITE (*,11) RSQ
11 FORMAT (5X,'RSQ= ',E13.6)
  IF (RSQ .LT. EPS) GO TO 33
  F(1)=P(1)*(A(1)+2*A(2)+2*A(3))/(B(1)+2*B(2)+2*B(3))
  F(2)=P(2)*(A(1)+2*A(2)+A(3)+A(4))/(B(1)+2*B(2)+B(3)+B(4))
  DO 20 IB=3,ND4
    J=IB-2
    K=IB+2
    TH=0.0
    TH1=0.0
    DO 15 II=J,K
      TH=TH+A(II)
      TH1=TH1+B(II)
15  CONTINUE
    IF (TH1 .EQ. 0.0) GO TO 20
    F(IB)=F(IB)*TH/TH1
20  CONTINUE
  DO 25 IC=2,ND2
    J=ND2+IC
    K=ND2+2-IC
    F(J)=F(K)

```



```

25 CONTINUE
30 CONTINUE
33 CONTINUE
  I=I-1
  WRITE (*,35) I
35 FORMAT (5X,'THE NO. OF ITERATIONS IS ',I3)
  RETURN
  END

```

C

```

SUBROUTINE PARAMS (N,A)
DOUBLE PRECISION A(2048),X(12),M(10),V,W
CHARACTER*7 NAME
K=1024
DO 20 I=1,12
  X(I)=0.D0
  DO 10 J=1,5
    K=K+1
    X(I)=X(I)+A(K)
10  CONTINUE
20 CONTINUE
  V=0.D0
  DO 30 I=1025,1084
    V=V+A(I)
30 CONTINUE
  DO 40 I=1,12
    X(I)=X(I)/V
40 CONTINUE
  DO 60 I=2,10,2
    M(I)=0.D0
    DO 50 J=1026,1084
      V=FLOAT(J-1025)
      W=FLOAT(I)
      V=V**W
      M(I)=M(I)+A(J)*V
50  CONTINUE
    M(I)=M(I)/59.D0
60 CONTINUE
  M(2)=DSQRT(M(2))
  V=M(2)
  DO 65 I=4,10,2
    M(I)=M(I)/(V**I)
65 CONTINUE
  PRINT*,'ENTER THE FILENAME FOR THIS SET OF'
  PRINT*,'PARAMETERS.'
  READ (*, '(A7)') NAME
  OPEN (90, FILE=NAME, PAD='YES')
  WRITE (90,70)
70 FORMAT (5X,'EVEN CENTRAL MOMENTS')
  WRITE (90,80) (I,M(I), I=2,8,2 )
80 FORMAT (10X,'M(',I1,') =',D17.10)
  WRITE (90,100) (M(10))

```

```

100 FORMAT (9X,'M(10) =',D17.10)
    WRITE (90,110)
110 FORMAT (5X,'5 CHANNEL GROUPS (CENTER FIRST)')
    WRITE (90,120) (I,X(I),I=1,9)
120 FORMAT (10X,'X(',I1,') =',D17.10)
    WRITE (90,130) (I,X(I),I=10,12)
130 FORMAT (9X,'X(',I2,') =',D17.10)
    CLOSE (90)
    RETURN
    END

```

C

```

    SUBROUTINE BACK (A,IMAX,N)
    DOUBLE PRECISION A(1024),C(2048),V
    COMPLEX*16 B(4096)
    CHARACTER*7 NAME
    ND2=N/2
    ND4=N/4
    DO 10 I=1,N
        C(I)=0.D0
10 CONTINUE
    N1=ND2-IMAX
    IF (N1 .GT. ND4) N1=ND4
    DO 20 I=1,N1
        J=IMAX+I-1
        C(I)=A(J)
20 CONTINUE
    N2=IMAX-1
    N3=N-N2
    DO 30 J=1,N2
        I=N3+J
        C(I)=A(J)
30 CONTINUE
    DO 40 I=1,N
        B(I)=DCMPLX(C(I),0.D0)
40 CONTINUE
    M=11
    CALL REORDER (M,B)
    CALL FFT (M,B)
    PRINT*,'WOULD YOU LIKE TO SEE THE FIRST FEW'
    PRINT*,'FOURIER COEFFICIENTS? '
    READ (*,'(A1)') NAME
    IF (NAME .EQ. 'N') GO TO 43
    PRINT*,'HOW MANY? '
    READ (*,*) IFC
    WRITE (*,41) (I,B(I),I=1,IFC)
41 FORMAT (I4,2D22.10)
43 CONTINUE
    PRINT*,'WOULD YOU LIKE TO SEE THE FIRST FEW'
    PRINT*,'VALUES OF THE POWER SPECTRUM? '
    READ (*,'(A1)') NAME
    IF (NAME .EQ. 'N') GO TO 46

```

```

PRINT*, 'HOW MANY? '
READ (*,*) IPS
DO 44 I=1,IPS
    A(I)=B(I)*CONJG(B(I))
44 CONTINUE
WRITE (*,45) (I,A(I),I=1,IPS)
45 FORMAT (I4,D22.10)
46 CONTINUE
PRINT*, 'DO YOU WISH TO DELETE ALL OF THE SINE'
PRINT*, 'COEFFICIENTS? '
READ (*, '(A1)') NAME
IF (NAME .EQ. 'N') GO TO 51
DO 50 I=1,ND2
    B(I)=(B(I)+CONJG(B(I)))/2.D0
50 CONTINUE
51 PRINT*, 'DO YOU WISH TO DELETE SOME OF THE COSINE'
PRINT*, 'COEFFICIENTS? '
READ (*, '(A1)') NAME
IF (NAME .EQ. 'N') GO TO 53
PRINT*, 'HOW MANY? '
READ (*,*) IDC
DO 52 I=1, IDC
    B(I)=(B(I)-CONJG(B(I)))/2.D0
52 CONTINUE
DO 152 I=2, IDC
    J=2050-I
    B(J)=(B(J)-CONJG(B(J)))/2.D0
152 CONTINUE
53 PRINT*, 'INSTEAD OF DELETING SOME OF THE COSINE'
PRINT*, 'COEFS. YOU MAY PREFER TO JUST TRIM THEM.'
PRINT*, 'THIS IS DONE BY SETTING THE FIRST N TERMS'
PRINT*, 'EQUAL TO THE N+1 TERM. YOU MAY INVOKE'
PRINT*, 'THIS OPTION EVEN IF YOU HAVE PREVIOUSLY'
PRINT*, 'DELETED THE FIRST N COSINE COEFFICIENTS.'
PRINT*, 'DO YOU WISH TO DO SO? '
READ (*, '(A1)') NAME
IF (NAME .EQ. 'N') GO TO 200
PRINT*, 'HOW MANY? '
READ (*,*) IT
DO 190 I=1, IT
    J=IT+1
    B(I)=DCMPLX(REAL(B(J)),0.D0)
190 CONTINUE
200 CONTINUE
PRINT*, 'DO YOU WISH TO DELETE SOME OF THE SINE'
PRINT*, 'COEFFICIENTS? '
READ (*, '(A1)') NAME
IF (NAME .EQ. 'N') GO TO 55
PRINT*, 'HOW MANY? '
READ (*,*) IDS
DO 54 I=1, IDS

```

```

      B(I)=(B(I)+CONJG(B(I)))/2.D0
54  CONTINUE
      DO 154 I=2,IDS
          J=2050-I
          B(J)=(B(J)+CONJG(B(J)))/2.D0
154  CONTINUE
55  CONTINUE
      PRINT*,'DO YOU WISH TO SAVE THE FOURIER COEFS. OF'
      PRINT*,'H(X) PRIOR TO THE TAPER IN THE WINGS? '
      READ (*,'(A1)') NAME
      IF (NAME.EQ. 'N') GO TO 57
      PRINT*,'ENTER THE FILENAME. '
      PRINT*,' '
      READ (*,'(A7)') NAME
      OPEN (90,FILE=NAME,PAD='YES')
      WRITE (90,56) (I,B(I),I=1,2048)
56  FORMAT (2(I4,2D22.10))
      CLOSE (90)
57  CALL REORDER (M,B)
      CALL FFT (M,B)
      DO 60 I=1,ND2
          C(I)=REAL(B(I))
60  CONTINUE
      V=0.D0
      DO 70 I=359,484
          V=V+C(I)
70  CONTINUE
      V=V/126.
      PRINT*,'DO YOU WANT A D.C. CORRECTION USING A'
      PRINT*,'LEAST SQUARE FITTING IN THE TAILS? '
      READ (*,'(A1)') NAME
      IF (NAME.EQ. 'N') V=0.D0
      DO 80 I=1,ND2
          C(I)=C(I)-V
          IF (C(I) .LT. 0.D0) C(I)=0.D0
80  CONTINUE
      DO 90 I=1,ND4
          J=(3*ND4)+I
          A(I)=C(J)
90  CONTINUE
      N4=ND4+1
      DO 100 I=N4,ND2
          J=I-ND4
          A(I)=C(J)
100 CONTINUE
      IMAX=513
      RETURN
      END

```

C

```

SUBROUTINE NOBACK (N,B,IMAX)
DOUBLE PRECISION B(1024),A(2048),V

```

```

      COMPLEX*16 C(4096)
      CHARACTER*7 NAME
      M=11
      ND2=N/2
      ND4=N/4
      DO 10 I=1,N
        A(I)=0.D0
10    CONTINUE
      N1=ND2-IMAX
      IF (N1 .GT. ND4) N1=ND4
      DO 20 I=1,N1
        J=IMAX+I-1
        A(I)=B(J)
20    CONTINUE
      N2=IMAX-1
      N3=N-N2
      DO 30 J=1,N2
        I=N3+J
        A(I)=B(J)
30    CONTINUE
      V=0.D0
      DO 33 I=1,N
        V=V+A(I)
33    CONTINUE
      DO 36 I=1,N
        A(I)=A(I)/V
36    CONTINUE
      OPEN (90,FILE='RAWDATA',PAD='YES')
      WRITE (90,70) (I,A(I),I=1,N)
      CLOSE (90)
      DO 40 I=1,N
        C(I)=DCMPLX(A(I),0.D0)
40    CONTINUE
      CALL REORDER (M,C)
      CALL FFT (M,C)
      DO 50 I=1,N
        A(I)=C(I)*CONJG(C(I))
50    CONTINUE
      PRINT*,'ENTER THE FILENAME FOR E(X) . '
      PRINT*,' '
      READ (*,'(A7)') NAME
      OPEN (90,FILE=NAME,PAD='YES')
      WRITE (90,60) (I,C(I),I=1,2048)
60    FORMAT (2(I4,2D22.10))
      CLOSE (90)
      PRINT*,'ENTER THE FLENAME FOR THE P.S. '
      PRINT*,' '
      READ (*,'(A7)') NAME
      OPEN (90,FILE=NAME,PAD='YES')
      WRITE (90,70) (I,A(I),I=1,1024)
70    FORMAT (5(I4,D22.10))

```



## APPENDIX G

In this Appendix the theoretical relationship between the vacancy concentration,  $C_v$ , and the concentration of dislocation cores,  $C_d$ , is demonstrated. (from Cotterill et al. 1972). The starting equation is

$$C_v = A \exp(-E_{vf}/KT) \quad (G.1)$$

where  $C_v$  is the vacancy concentration at thermal equilibrium

$E_{vf}$  is the vacancy formation energy

and  $KT$  has its usual meaning

Now the concentration of vacancies after the quench,  $C'_v$ , is related to  $C_v$  by

$$C'_v = nC_v \quad (G.2)$$

where  $n$  is the quench efficiency which is assumed to be nearly unity

After the low temperature anneal the concentration of dislocation loops,  $C_L$ , is given by

$$C_L = \nu C'_v \quad (G.3)$$

where  $\nu$  is the inverse of the number of vacancies required to form an average loop

The dislocation concentration,  $C_d$ , in terms of an atomic fraction is then

$$C_d = C_L \pi D / d_0 \quad (G.4)$$

where  $D$  is the average loop diameter

and  $d_0$  is the nearest neighbor distance.

It can be shown that

$$v = 2\sqrt{3} d_o^2 / \pi D^2 \quad (G.5)$$

Combining Equations (G.3), (G.4), and (G.5) yields

$$C_d = 2\sqrt{3} d_o C_v' / D \quad (G.6)$$



## APPENDIX H

In this Appendix the derivation of the defect specific lineshape parameter "R" is recreated from the work of Mantl and Trifthauser (1978). The following quantities require definition:

- $S, W$  - are the values of the "S" and "W" parameters for the sample being analyzed.
- $S_t, W_t$  - are the values of the "S" and "W" parameters for a sample in which all of the positrons are annihilating from the trapped state.
- $S_B, W_B$  - are the values of the "S" and "W" parameters for a sample in which all of the positrons are annihilated from the bulk (defect free).
- $P_t$  - the probability of a positron annihilation from the trapped state.
- $P_B$  - the probability of a positron annihilating from the bulk.
- $\mu$  - the trapping constant.
- $C$  - the trap concentration.
- $\lambda_B$  - the annihilation rate from the bulk state.

Now, it is known that

$$S = S_B P_B + S_t P_t \quad (H.1)$$

and

$$P_B + P_t = 1 \quad (H.2)$$

Also

$$P_t = \mu C / (\lambda_B + \mu C)$$

(H.3)

Substituting Eq. (H.3) into Eq. (H.1) yields:

$$S = S_B \left(1 - \frac{\mu C}{\lambda_f + \mu C}\right) + S_t \left(\frac{\mu C}{\lambda_f + \mu C}\right) \quad (H.4)$$

Similarly

$$W = W_b \left(1 - \frac{\mu C}{\lambda_f + \mu C}\right) + W_t \left(\frac{\mu C}{\lambda_f + \mu C}\right) \quad (H.5)$$

Rearranging Eq. (H.4) and Eq. (H.5) yields:

$$S - S_B = (S_t - S_B) \mu C / (\lambda_f + \mu C) \quad (H.6)$$

$$W - W_B = (W_t - W_B) \mu C / (\lambda_f + \mu C) \quad (H.7)$$

Taking the ratio of Eq. (H.6) to Eq. (H.7) yields:

$$R = \left| \frac{S - S_B}{W - W_B} \right| = \left| \frac{S_t - S_B}{W_t - W_B} \right| \quad (H.8)$$

Clearly the ratio on the far right has a constant value for a given defect in a given material.

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## BIOGRAPHY

James Peter Schaffer was born on November 13, 1959, in Wilmington, Delaware. He received his Bachelor of Science in Mechanical Engineering and Materials Science (Magna Cum Laude) in 1981 from Duke University in Durham, North Carolina. He received his Master of Science degree in Mechanical Engineering and Materials Science in the following year.

Mr. Schaffer published several papers during his years as a graduate student. These include:

- 1) "Photoconductivity of Amorphous Hydrogenated Silicon" with J. H. Lind, P. L. Jones, and F. H. Cocks, Physica Status Solidi (a) 71, K111, 1982.
- 2) "Hydrogenated Amorphous Boron: Transient and Steady State Photoconductivity" with H. Park, J. H. Lind and P. L. Jones, Physica Status Solidi (a) 81, K51, 1984.
- 3) "The Deconvolution of Doppler Broadened Positron Annihilation Measurements using Fast Fourier Transforms and Power Spectral Analysis" with E. J. Shaughnessy and P. L. Jones, Nuclear Instruments and Methods in Physics Research B5, 75, 1984.
- 4) "A Comparison of the Doppler Broadened Positron Annihilation Spectra of Neutron Irradiated  $\text{Al}_2\text{O}_3$  and  $\text{MgAl}_2\text{O}_4$ " with P. L. Jones, F. H. Cocks, F. W. Clinard, Jr., and G. F. Hurley, Journal of Nuclear Materials 127, 221, 1985.

In addition, Mr. Schaffer presented a paper entitled "On the Application of Fast Fourier Transforms and Power Spectrum Analysis to Incorrectly Posed Integral Equations" at the Integral Methods in Science and Engineering Conference in Arlington, Texas, March 18-21, 1985.

In 1981, Mr. Schaffer received a research and teaching assistantship award from Duke University for graduate study in the Department of Mechanical Engineering and Materials Science. In the following year he was awarded a three year Fellowship from the Office of Naval Research. During his graduate studies, Mr. Schaffer was inducted into Sigma Xi, the National Scientific Research Society.

## A positron annihilation lifetime spectroscopy study of porous silicon using a continuous lifetime fitting algorithm

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Porous silicon is a highly complex system containing disorder that exists over a range of scales. Depending on the anodization current and time, and the dopant level and type, the resulting porous layer can have a porosity of up to 80% with a unidirectional columnar-like structure for  $p^-$  silicon wafers [1]. The remaining silicon varies in size with characteristic lengths ranging between from 3–4 to 15–30 nm. Such small crystalline structures have an enlarged bandgap due to the quantum confinement effect. The role of confinement in the observed increase in radiative inter-band transition efficiency is however still unclear. How the broken silicon bonds are terminated can directly affect the nature of the inter-band transitions. For example, hydrogen passivation "sweeps" the dangling bond states out of the band gap region; reducing the available non-radiative recombination paths and thereby increasing the efficiency of the radiative transitions. This, however, is not the complete picture since the oxygen content of the porous region has been found in some cases to amplify and change the photoluminescence properties [2]. The electro-chemical processes occurring on the surface during anodization are not yet clearly understood and further experimental characterization is needed.

Positron annihilation lifetime spectroscopy (PALS) provides a unique non-destructive probe into the disorder inherent in porous silicon on the microscopic scale, through the identification of impurity concentrations and types; and on the mesoscopic scale at which the bulk of the porosity exists through the study of positronium lifetimes. Itoh *et al.* [3,4] and Dannefaer *et al.* [5] have done such an investigation using a discrete lifetime analysis. Itoh *et al.* obtained a satisfactory three-component fit, finding a bulk silicon component (at 230 ps), a defect component (at 600 ps), which they interpreted as arising from either silicon mono-vacancies and/or oxygen divacancies within an Si–O<sub>2</sub> layer [6], and a longer 25 ns lifetime due to ortho-positronium annihilating in the porous region. Dannefaer *et al.*, on the other hand, determined that a five-component fit was better, with additional lifetimes arising from para-positronium (at 125 ps) and another ortho-positronium component at 2.5 ns. They attributed the latter to ortho-positronium annihilating in larger vacancy defects either within the remaining silicon or the oxide layer. In this letter we report on a PALS investigation of porous silicon using a continuous lifetime fitting algorithm.

Our motivation lies in the underlying disadvantage in discrete lifetime fitting algorithms where the number of components must initially be assumed since in general a realistic spectrum does not uniquely determine this number. This becomes particularly apparent when looking at highly disordered systems where, indeed, the notion of a discrete spectrum may be invalid. These problems can in part be overcome by using a continuous lifetime fitting algorithm; for example, performing an inverse Laplace transform on the lifetime spectrum, thereby obtaining a probability density function of characteristic lifetimes. The Fortran program CONTIN [7–9] is such an algorithm. Admittedly, use of the inverse Laplace transform does not take into account the instrument resolution function of the apparatus. For the determination of sub-nanosecond lifetimes this entails a distinct loss of accuracy. For well-separated lifetimes that are significantly larger than the typical full width at half maximum (FWHM) of the equipment, the effect is reduced and overshadowed by the loss of accuracy due to the worsening statistics typical for these components. To date, our work appears to be the only systematic study of porous silicon using a continuous lifetime fitting algorithm. This is the first step towards achieving the goal of determining intrinsic lifetime distribution widths for positrons annihilating within the porous silicon layer, and thus its electronic structure.

For the measurement of the annihilation lifetime spectra of porous silicon we employed a standard PALS rig at 200 ps per channel. Such a large timing window is needed to ensure that all lifetime components decay completely into the background. The positron source was <sup>22</sup>NaCl evaporated onto a 6  $\mu$ m thick titanium film. Using our PALS rig at 50 ps per channel and the discrete lifetime fitting program PFPOSFIT [10], we obtained for bulk silicon,  $221 \pm 1$  ps at  $97 \pm 1\%$  concentration, with a source component lifetime of approximately  $429 \pm 25$  ps at  $3 \pm 1\%$  concentration (the iterated instrument resolution had an FWHM of 240 ps with long and short time tails of 50 and 20 ps, respectively).

The porous silicon samples for this investigation were made from 200  $\mu$ m  $p^-$  doped wafers, anodized for 30 min at 20 mA cm<sup>-2</sup>. The electrolyte used was a 35% ethanoic solution of HF. Under illumination by an ultraviolet (UV) lamp this sample glowed a deep red. The initial thickness of the porous layer



was approximately 50–60  $\mu\text{m}$ . A high resolution scanning electron microscopy (SEM) analysis performed approximately 6 months after the PALS experiment revealed a layer that was approximately 10  $\mu\text{m}$  thick, indicating that structural damage had occurred (Fig. 1). The sample nevertheless still glowed red at a reduced intensity. Fig. 1 reveals the large-scale columnar structure typical of  $p^-$  doped porous silicon samples; the remaining (porous) silicon constituting the much thinner connecting membranes between the micrometre-sized pores.

All PALS runs were taken at room temperature (24 °C) in an environment where the temperature could be held constant to within a degree. For each sample, a varying number of spectra were taken (see Table I and Fig. 2), each of which had a maximum peak height of  $10^6$  counts. Statistical analysis across the spectra for a particular sample indicated that to within the noise level, there was no detectable difference between individual runs, justifying the use of the mean spectrum in the analysis using CONTIN. The associated information content analyses [11, 12] are also given in Table I. Here, the Shannon number indicates that in principle up to eight distinct lifetime peaks are resolvable, and the resolution factor suggests that the minimum ratio between these resolvable lifetimes ranges between 2 and 3.

Since CONTIN assumes a spectrum constructed from exponentials, we must choose an appropriate zero-time channel from which the analysis can begin. This must be just beyond the peak of the spectrum, yet not too close to include the "rounding off" effect of the finite instrument resolution. The resulting inverse Laplace transform found by CONTIN is not very sensitive to the choice of the zero-time channel as far as determining distinct components. It is, however, sensitive to determining their

intensity, since a shift  $\delta t$  in the zero-time channel will affect the intensity of a component  $\tau$  by the approximate factor  $\exp(-\delta t/\tau)$ .

Such an analysis for the 200 ps per channel bulk silicon wafer spectrum is displayed in Fig. 2 for two different zero-time channels; approximately 400 ps (solution 1) and 800 ps (solution 2) beyond the instrumental zero-time channel. Solution 1 contains a dominant broad component centred at approximately 220 ps, which can be attributed to the bulk silicon component. Two long time components can be seen, occurring at approximately 1.5 and 45 ns, the shorter of which can be associated with the common ortho-positronium "pick off" signal present in semiconductors [13]. The extra long 45 ns component does not seem to have been encountered in the literature. We do not know at this stage the origin of this lifetime peak; however, as we shall see, it also appears in the porous silicon sample at an increased intensity. In solution 2, the bulk silicon component becomes broader and less intense, the ratio of its peak height with that of solution 1 being approximately  $\exp(\delta t/220)$  where  $\delta t \approx 400$  ps. The ortho-positronium peaks at 1.5 and 50 ns acquire more dominance, with their peak heights largely unchanged from solution 1. In addition, we observed a shorter, approximately 125 ps, component, which is most likely to be the accompanying para-positronium signal. That this was not resolved in solution 1 suggests that the "rounding off" effect was not completely removed at 400 ps beyond the approximate zero-time channel.

Fig. 3 displays the CONTIN solution for the porous silicon spectra. Again, solution 1 and solution 2 are for time shifts of approximately 400 and



Figure 1 High resolution SEM picture of the porous silicon sample used in the PALS investigation.

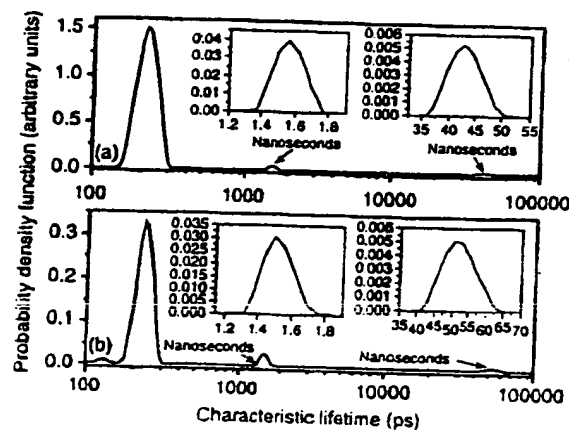


Figure 2 PALS lifetime spectra of (a) bulk silicon and (b) porous silicon taken at 200 ps per channel.

TABLE I The number of PALS spectra taken and the resulting resolution factor and Shannon number

Sample	Number of spectra	Resolution factor	Shannon number
Bulk silicon at 200 ps per channel	49	2	9
Porous silicon at 200 ps per channel	24	3	8

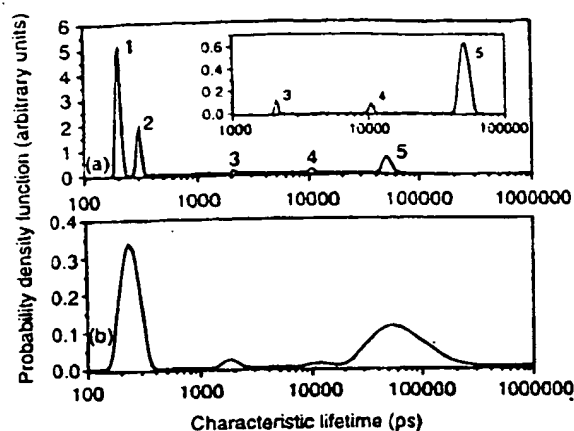


Figure 3 Characteristic lifetime probability density determined using CONTIN for the bulk silicon sample taken at 200 ps per channel. (a) Solution 1: zero-time approximately 400 ps beyond the instrumental zero-time channel. (b) Solution 2: zero-time approximately 800 ps beyond the instrumental zero-time channel.

800 ps beyond the instrumental zero-time channel, respectively. Inspection of solution 1 indicates five distinct components. Component 1 is centred at approximately 200 ps and can be attributed to the positrons annihilating either within the silicon substrate or in the remaining silicon in the porous material. Component 2 is centred at approximately 300 ps and has also been encountered by both Itoh *et al.* and Dannefaer *et al.* Their measured lifetimes for component 2, however, ranged between 400 and 600 ps. CONTIN analyses of simulated spectra obeying Poisson statistics indicate that this discrepancy (and the reduced bulk lifetime) may be due to the 200 ps per channel resolution and the neglect of the instrument resolution function. Components 3, 4 and 5, occurring respectively at approximately 2, 10 and 50 ns, are the long lifetime components due to ortho-positronium. In solution 2, the sub-nanosecond components are reduced in intensity, and crystalline silicon and defect components (at 300 ps) are no longer separately resolvable, merging into a single broad peak centred at approximately 250 ps. The long time components now become broader and of a reduced height. This indicates that the widths we have observed are not intrinsic to the material but rather an artefact of the fitting process and the signal to noise ratio. Marshall [14] has concluded that to determine accurately the intrinsic widths of peaks, this ratio must be at least 400. Inspection of Fig. 4 indicates a signal to noise ratio of less than 40 for the positronium decay modes.

The present method of analysis is unsuitable to obtain accurate absolute intensity information since the zero-time channel cannot be estimated beyond the 200 ps per channel limit. Furthermore, modifications to CONTIN suggested by Gregory [12, 15–17] that take into account the finite resolution (and therefore the zero-time channel), by obtaining the probability density profile with respect to a reference spectrum (in our case, bulk silicon), have proved unsuccessful. This is primarily due to the poor

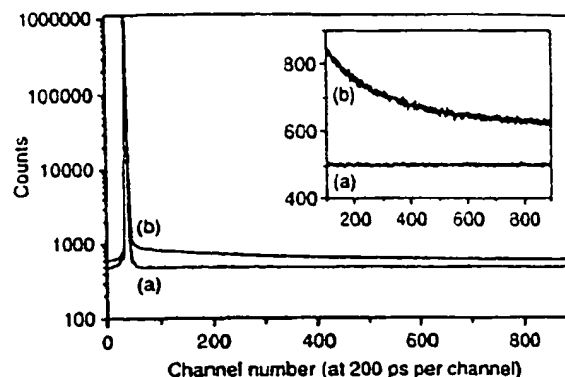


Figure 4 Characteristic lifetime probability density determined using CONTIN for the porous silicon sample taken at 200 ps per channel. (a) Solution 1: zero-time approximately 400 ps beyond the instrumental zero-time channel. (b) Solution 2: zero-time approximately 800 ps beyond the instrumental zero-time channel.

definition of the bulk silicon reference spectrum at 200 ps per channel.

Using the PALS data collected from sample A, we have confirmed and extended the findings of both Itoh *et al.* and Dannefaer *et al.* In particular, we resolved three rather than two ortho-positronium components, suggesting that there may be an additional intermediary scale of porosity in which ortho-positronium annihilates. This has a lifetime indicative of hydrogenated amorphous silicon [18], suggesting ortho-positronium annihilation again within the amorphous layer. The integrated areas (corrected for zero-time shift) of the positronium peaks in solutions 1 and 2 show that components 3 and 5 occur in approximately the same proportion; Dannefaer *et al.*, on the other hand, found that the ~2.5 ns component occurred an order of magnitude less than the longest time ortho-positronium component. The present analysis also establishes the existence of a very weak ortho-positronium component in the pre-anodized wafers at a time scale approximately equal to the longest time ortho-positronium component seen in porous silicon, using theory [19] that has traditionally been employed in the calculation of free volumes in polymer materials. In porous silicon Itoh *et al.* associated a void length scale of approximately 1 nm to this ortho-positronium component, suggesting that irregularities of this magnitude exist on the pre-anodized wafer and that these may, in part, be the catalyst for the initial pore formation process.

The results presented here further demonstrate the rich variety of disorder inherent in porous silicon and indicate the potential of a continuous lifetime analysis of PALS spectra to provide a high resolution probe into such structures. If another order of magnitude increase in signal to noise ratio can be achieved then there exists the possibility of resolving intrinsic peak widths for porous silicon samples, thereby extending the PALS method to resolving pore size distributions using the ortho-positronium components, and conceivably giving insight into the electronic structure of the confined electrons within the remaining silicon crystallites of the porous layer.

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# Study of the Van Cittert and Gold iterative methods of deconvolution and their application in the deconvolution of experimental spectra of positron annihilation

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## Abstract

The study of deconvolution by Van Cittert and Gold iterative algorithms and their use in the processing of experimental spectra of Doppler broadening of the annihilation line in positron annihilation measurement is described. By comparing results from both algorithms it was observed that the Gold algorithm was able to eliminate linear instability of the measuring equipment if one uses the 1274 keV  $^{22}\text{Na}$  peak, that was measured simultaneously with the annihilation peak, for deconvolution of annihilation peak 511 keV. This permitted the measurement of small changes of the annihilation peak (e.g.  $S$ -parameter) with high confidence. The dependence of  $\gamma$ -ray-like peak parameters on the number of iterations and the ability of these algorithms to distinguish a  $\gamma$ -ray doublet with different intensities and positions were also studied.

## 1. Introduction

Positron annihilation (PA) as one of the non-destructive nuclear methods is used for the study of structure of different materials and it is especially efficient in determining free volumes or defects in bulk material. The main characteristics, that are determined by PA, are lifetimes of positrons in matter, Doppler broadening of annihilation line (DBAL), and angular correlation of annihilation  $\gamma$ -rays. The last two characteristics give us information about the momentum distribution of electrons in matter. The more common angular correlation method is approximately one order more accurate, but it is more time consuming and requires a more intensive positron source. On the other hand DBAL is simple and not time consuming. It is mainly used for determining the relative changes in the annihilation peak (e.g.  $S$ -parameter, defined as the ratio between area of central part of the peak to the full area of the peak) depending on temperature, pressure etc. After deconvolution of the annihilation peak using a resolution function of measuring system, one can obtain the momentum distribution of electrons in the sample. If we are only interested in relative changes of the  $S$ -parameter, deconvolution is not necessary. But as we have seen in our work, deconvolution can partly eliminate the spurious effects due to instability of the measuring system. Another possibility to eliminate an instability of the system is the use of an expensive spectrum stabilizer unit.

Generally, deconvolution algorithms have recently found many applications in various domains of experimental science. They have been applied to such different problems as improving resolution in spectroscopy and measuring the thickness of multilayer structures. There is also the long-standing problem of determination of position and intensities in  $\gamma$ -ray multiplets [1].

## 2. Theory

The relationship between a measured value  $x(t)$  and the raw result of measurement  $y(t)$  can be described by a convolution-type integral equation

$$y(t) = \int_{-\infty}^{\infty} x(\tau) h(t - \tau) d\tau, \quad (1)$$

where  $h(t)$  is an impulse response. Knowledge of the instrumental function  $h(t)$  is usually required. For a discrete system, Eq. (1) can be written as

$$y(i) = \sum_{k=0}^{N-1} h(i-k)x(k) \quad i = 0, 1, \dots, 2N-2, \quad (2)$$

where  $N$  is the number of samples of vectors  $h, x$ .

The impulse response has a finite length. Therefore we will assume that  $h(i) = 0$  for  $i < 0$  and  $i \geq N$ . Then Eq. (2) can be written in a matrix form:

$$\begin{bmatrix} y(0) \\ y(1) \\ y(2) \\ \vdots \\ \vdots \\ \vdots \\ y(2N-2) \end{bmatrix} = \begin{bmatrix} h(0) & 0 & 0 & \dots \\ h(1) & h(0) & 0 & \dots \\ h(2) & h(1) & h(0) & \dots \\ \vdots & \vdots & \vdots & \ddots \\ h(N-1) & h(N-2) & h(N-3) & \dots \\ 0 & h(N-1) & h(N-2) & \dots \\ 0 & 0 & h(N-1) & \dots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix} \begin{bmatrix} x(0) \\ x(1) \\ x(2) \\ \vdots \\ \vdots \\ x(N-1) \end{bmatrix}, \quad (3)$$

or

$$y = Hx. \quad (4)$$

It means that the columns of  $H$  are represented by vectors  $h$  mutually shifted by one position. Multiplying both sides of Eq. (4) by  $H^T$  gives

$$H^T y = H^T H x, \quad (5)$$

or

$$y_1 = H_1 x, \quad (6)$$

where  $H_1$  is Toeplitz matrix [2,3].

Solution of the linear equation system (6) (vector  $x$ ), under the condition that the output vector  $y$  and the matrix of the impulse response  $H$  is known, is a problem of deconvolution. The output vector of system  $y$  is affected by noise that accompanies each measurement. The existence of this noise strongly affects the process of deconvolution, and can lead to difficulties in solving the linear equation system (6).

The above-formulated problem of input reconstruction is as a rule ill-conditioned, i.e. the estimates  $\hat{x}(t)$  of  $x(t)$  satisfying Eq. (1) are extremely sensitive to errors in the measured data  $y(t)$ . It is expressed by the fact, that the matrix  $H_1$  is almost singular. The direct inversion of  $H_1$  for solving  $x$  cannot lead to a stable solution.

Therefore, in order to solve this problem, the method of regularization must be included. This means that original problem is replaced by an approximate one which solutions are significantly less sensitive to errors in the data  $y(t)$ .

The Van Cittert iterative method of deconvolution is widely applied in different areas, for example in spectroscopy or in image processing [2,4]. The Van Cittert algorithm of deconvolution is described in detail in Ref. [4], so we will describe it only very briefly.

Its basic form for a general linear discrete system is

$$x^{k+1} = x^k + \mu(y - Ax^k), \quad (7)$$

where  $A$  is system matrix,  $k$  represents the number of iterations and  $\mu$  is the relaxation factor. The convergence condition of Eq. (7) is that the diagonal elements of the matrix  $A$  satisfy

$$A_{ii} > \sum_{j=0, j \neq i}^{N-1} A_{ij}, \quad i = 0, 1, \dots, N-1. \quad (8)$$

It is obvious that such a diagonal element dominance is rare in physical problems. However, the deconvolution algorithm in Eq. (7) can be modified in a such way that it will satisfy the conditions of convergence. Hence, Eq. (7) becomes

$$x^{k+1} = \mu y + (E - \mu A)x^k = \mu y + Dx^k, \quad (9)$$

where  $E$  is a unit matrix and

$$D = E - \mu A. \quad (10)$$

Under the condition that  $x^0 = \mu$ , the successive substitutions give

$$x^k = \mu y + \mu D y + \dots + \mu D^{k-1} y + \mu D^k y = \mu(E + D + \dots + D^k)y. \quad (11)$$

Assuming that  $\lambda_0, \lambda_1, \dots, \lambda_{N-1}$  are eigenvalues of  $A$ , then  $(1 - \mu\lambda_0), (1 - \mu\lambda_1), \dots, (1 - \mu\lambda_{N-1})$  are eigenvalues of  $D$ . If

$$\lim_{k \rightarrow \infty} (1 - \mu\lambda_i)^k = 0, \quad i = 0, 1, \dots, N-1, \quad (12)$$

then

$$\lim_{k \rightarrow \infty} D^k = [0],$$

and

$$\lim_{k \rightarrow \infty} x^k = A^{-1}y = x. \quad (13)$$

From equation (12) this implies that the necessary and sufficient conditions of convergence are

$$|1 - \mu\lambda_i| < 1, \quad i = 0, 1, \dots, N-1. \quad (14)$$

If we define  $\lambda_i$  and its conjugate  $\lambda_i^*$  as

$$\lambda_i = a_i + jb_i, \quad \lambda_i^* = a_i - jb_i,$$

then the convergence condition (14) becomes

$$\mu[\mu(a_i^2 + b_i^2) - 2a_i] < 0, \quad i = 0, 1, \dots, N-1. \quad (15)$$

Inequality (15) gives two bounds for  $\mu$

$$\mu = 0, \quad \mu = 2 \frac{a_i}{a_i^2 + b_i^2}, \quad i = 0, 1, \dots, N-1. \quad (16)$$

$N$  conditions determine the bounds of the  $\mu$  coefficient. Unfortunately these conditions are not fulfilled for all cases. However, if the system matrix  $A$  is positive definite the convergent solution always exists. So we settle the algorithm in a such way that the eigenvalues  $\lambda_i$  will be positive, real numbers.

Let us return to Eq. (5). Matrix  $H^T H$  is symmetric, so its eigenvalues are real. The eigenvalues of matrix  $(H^T H)(H^T H)$  are squares of eigenvalues of matrix  $H^T H$  and therefore must be positive. Eq. (5) becomes

$$(H^T H H^T)y = (H^T H H^T)x, \quad (17)$$

and the iterative algorithm of deconvolution becomes

$$x^{k+1} = x^k + \mu[(H^T H H^T) y - (H^T H H^T H) x^k], \quad (18)$$

or

$$x^{k+1} = x^k + \mu[y' - H' x^k]. \quad (19)$$

By this we ensure the existence of a common interval of solution for inequality (14) and convergence of the deconvolution algorithm. Eigenvalues  $\lambda_i$  are real, positive numbers, so from Eq. (16) for  $\mu$  we can write

$$0 < \mu < \frac{2}{\lambda_{\max}}, \quad (20)$$

where  $\lambda_{\max}$  is the greatest eigenvalue of  $H'$

$$\lambda_{\max} = \max(\lambda_0, \lambda_1, \dots, \lambda_{N-1}). \quad (21)$$

Now we determine the maximum eigenvalue  $\lambda_{\max}$ . For eigenvalues of system (17) we can write

$$H' x = \lambda_i x, \quad i = 0, 1, \dots, N-1. \quad (22)$$

If  $x_j$  is the biggest absolute element in  $x$ , then from Eq. (22) we get

$$\sum_{m=0}^{N-1} H'_{jm} x_m = \lambda_i x_j \quad (23)$$

or

$$\lambda_i = \frac{\sum_{m=0}^{N-1} H'_{jm} x_m}{x_j}. \quad (24)$$

Then

$$\lambda_i \leq \sum_{m=0}^{N-1} |H'_{jm}|, \quad i = 0, 1, \dots, N-1. \quad (25)$$

In practical cases we do not know the biggest element in  $x$ . We determine the value of  $\lambda_{\max}$  as the maximum value from  $\lambda_i$ , determined by Eq. (25) e.g., from the sum of absolute values of rows in matrix  $H'$ .

This is the base of the Van Cittert algorithm of deconvolution. Now we introduce, in analogy with Eq. (24), a local variable relaxation factor

$$\mu_i = \frac{x_i^k}{\sum_{m=0}^{N-1} H'_{im} x_m^k}, \quad (26)$$

and we use it in Eq. (19). For the  $i$ th element of vector  $x^{k+1}$  we get

$$x_i^{k+1} = x_i^k + \frac{x_i^k}{\sum_{m=0}^{N-1} H'_{im} x_m^k} \left[ y_i - \sum_{m=0}^{N-1} H'_{im} x_m^k \right]. \quad (27)$$

or

$$x_i^{k+1} = \frac{y_i}{N-1} \frac{x_i^k}{\sum_{m=0}^N H_{im} x_m^k} \quad (28)$$

Eq. (28) is the Gold algorithm of deconvolution [5]. It is an extension of Van Cittert's iterative method. The advantage of Gold's method is that the solution  $x$  is positive.

### 3. Theoretical simulation

To test an applicability of Van Cittert's and Gold's methods of deconvolution for  $\gamma$ -rays spectra we performed a theoretical simulation. We convoluted the experimental resolution function (RF) obtained from a positron annihilation experiments with different kinds of Gaussians describing  $\gamma$ -ray peaks. The simulated spectra were used as an input to both Gold's and Van Cittert's iterative deconvolution procedures. Simulated spectra included a noise component due to an experimental resolution function.

#### 3.1. The dependence of output peak parameters from the deconvolution on the number of iterations

Because the deconvolutions are using the iterative method, it was interesting to study the dependence of the output from the deconvolution on the number of iterations ( $N$ ).  $N$  was varied in the range 200-50 000. 2000 iteration steps lasted 40 min on a PC486, 33 MHz for 200 channels of the spectrum (a version with elimination of the multiplication of zero matrix terms took approx. 15 min).

We analyzed the peak which was a convolution of a Gaussian with FWHM = 13 channels and a resolution function with a FWHM = 10.4. The resulting peak had a FWHM = 16.7, which was a value in the range of our experimental values for the FWHM of annihilation peaks measured in DBAL. The deconvoluted peak was fitted and the result should be one Gaussian with a FWHM = 13.

Fig. 1 shows the dependences of the  $\chi^2$  value and FWHM on the number of iterations for the algorithm of Van Cittert and Gold. As we can see from this figure, the resulting deconvolution fit for the Van Cittert algorithm was much better for two Gaussians than for one until 14 000 iterations, then the  $\chi^2$  became practically the same.

For the Gold algorithm the  $\chi^2$  of the deconvolution fit was very good for one Gaussian and reached a minimum value for 2000 iterations. The increase of  $\chi^2$  with the number of iterations (for both Van Cittert and Gold methods) is a consequence of the round-off errors which arise during the computations with real numbers represented in standard single precision (REAL in Fortran or FLOAT in C) and a large number of these

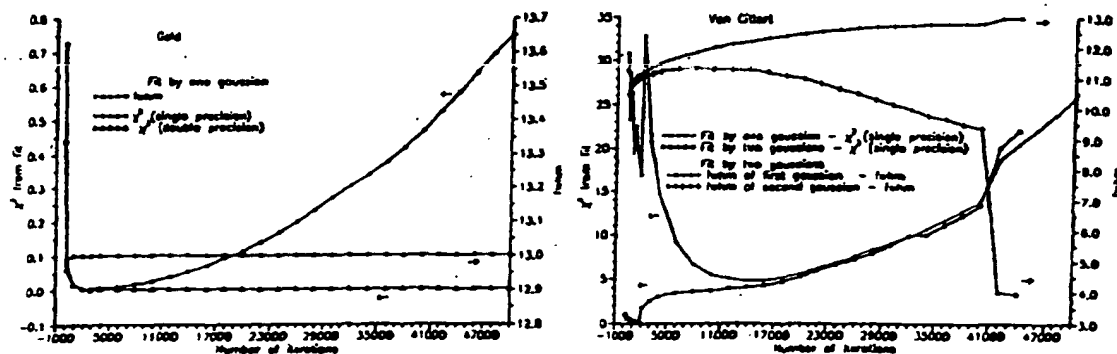


Fig. 1. Dependence of  $\chi^2$  and FWHM from fit of deconvoluted peak on number of iteration steps.



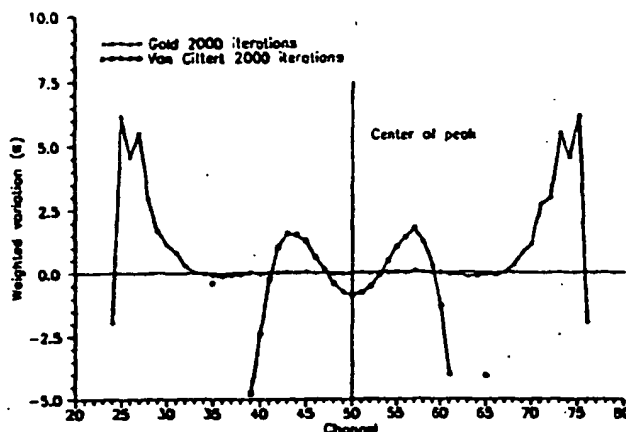


Fig. 2. Weighted variations of deconvoluted peak and original Gaussian.

operations. It was verified by a calculation (for the Gold method) using the double precision floating-point representation of real numbers. The results of the double-precision calculation are shown in Fig. 1. The fast convergence and the stability of the solution can be observed. It clearly shows the possible errors arising from using a finite computer word length of and a large number of iteration steps.

For the Van Cittert algorithm, it can be seen that by increasing the number of iterations the first Gaussian became more and more dominant and approached the input Gaussian in convolution. The same dependence can be observed in the intensity of Gaussians.

For the Gold algorithm, the results were much better. From Fig. 1, it is clear that the FWHM from the fit of the deconvoluted peak reached the correct value of 13 for the number of iterations larger than 1000. The variance of the FWHM from this fit was  $< 0.03\%$  and of the amplitude  $< 0.005\%$  for  $N \geq 1000$ .

For the Van Cittert algorithm and  $N \geq 2000$ , the variance of the FWHM from the fit was  $< 0.3\%$  and of the amplitude  $< 0.05\%$ .

In Fig. 2 the weighted differences of the original input Gaussian and fitted results of the deconvoluted peak after  $N = 2000$  iteration steps are shown. The difference is evident. For the Gold deconvolution the variance was still less than 7.5% even for the  $\pm 25$  displaced channels from the center of the peak. For the large number of iterations ( $N > 30000$ ) there was already deformation in deconvoluted peak, which could be seen as sharp peaks in the dependence of weighted differences. This deformation is a consequence of round-off errors in the computer.

Fig. 3 shows typical results of the deconvolution procedures. The peak, deconvoluted by the Gold's algorithm, is almost identical with the original Gaussian after carrying out 2000 iterations. Van Cittert's procedure showed negative values and there were oscillations on both wings of the peak on the level of 2% of amplitude of peak. Because Fig. 3 is in a logarithm scale the negative values are not displayed. Gold's deconvolution did not produce negative values and oscillations were very small ( $< 0.01\%$ ).

### 3.2. The deconvolution of Gaussians with different FWHM

To determine the influence of the FWHM of the convoluted Gaussian on the result of deconvolution, we also made a convolution of the Gaussian with  $\text{FWHM} = 20$  with RF. Then the difference in parameters of the resulting deconvoluted peak after 2000 iterations was compared with the difference for  $\text{FWHM} = 13$ .

The Van Cittert's deconvolution of convolution of the original Gaussian with a  $\text{FWHM} = 20$  was in very good agreement with the original Gaussian,  $\chi^2$  from the fit was 0.04 and the FWHM was exactly the same.

The weighted difference of the fit of deconvoluted and original Gaussian was  $\leq 10\%$  for counts  $> 0.1\%$  of amplitude. The situation was worse for the convolution of the Gaussian with  $\text{FWHM} = 13$ . The FWHM of the deconvoluted peak was larger than about 1.8%. But the worst fact was that the  $\chi^2$  from the fit was

very large (Fig. 1) and the deconvolution was better described by two Gaussians ( $\chi^2 = 1.8$ ). That means that the deconvolution changed the shape of the peak in such a way that the deconvoluted peak could be better described by two Gaussians though there should be only one. The weighted difference of the deconvolution fit and the original Gaussian was  $\leq 10\%$  for counts  $> 3\%$  of the amplitude. The parameters from the fit of deconvoluted peak were in excellent agreement with the original Gaussian. Problem was that for a Gaussian with FWHM = 13 there was a better  $\chi^2$  for two Gaussians as for one.

For the Gold deconvolution agreement with the original Gaussian was very good also for Gaussian with FWHM = 13 as we can see from Fig. 2. What was important was that there are no deformations in deconvoluted peak because the  $\chi^2$  for one Gaussian was very small (Fig. 2).

### 3.3. The deconvolution of the sum of two Gaussians with different intensity and position

To verify the possibility of distinguishing components of a doublet (sum of two peaks) by deconvolution, we made convolutions of the sum of two Gaussians with intensity ratios 1:1, 1:0.8, 1:0.5, 1:0.2 and mutual displacements of 1, 5, 10, 15, 20 channels. Both input Gaussians had FWHM = 13, the number of iteration steps  $N$  was 2000 and the deconvoluted peaks were fitted. We computed the weighted difference of these fitted parameters with the parameters of the original input Gaussians.

For the Van Cittert's deconvolution we got the following results:

If the peak displacement was 15 and 20 channels there was no problem to fit the deconvolution results. The variation from the parameters of the input Gaussians was small,  $\leq 2\%$ . For a displacement of about 10 channels, the fitting procedure for the ratio of intensities 1:0.2 failed. The variance was  $\leq 5\%$  for ratios 1:1, 1:0.8 and  $< 19\%$  for 1:0.5. For a displacement of about 5 channels we were not able to get a fit for 1:0.2 and 1:0.5 ratios. The variances were  $< 42\%$  (the largest for the amplitude, for the remaining parameters  $< 10\%$ ). So for peak displacements larger than the FWHM of the peaks, the resolution was good also for the ratio of intensity of 20%. For a displacement smaller as the FWHM of the peak, the resolution was possible only for intensities close to each other. For a small displacement ( $< 10\%$ ), the fit was impossible.

For the Gold deconvolution we also got a fit for a displacement of 10 channels and ratio 1:0.2 and a displacement of 5 channels and ratios 1:0.5, 1:0.2 with variances  $< 27\%$ . For a displacement of 1 channel the fitting procedure failed. So we can say that Gold's algorithm is much more able to distinguish a doublet and give us more precise results than the Van Cittert's algorithm.

In Fig. 4 convolutions of Gaussians and the results of the deconvolution procedure described above can be seen.

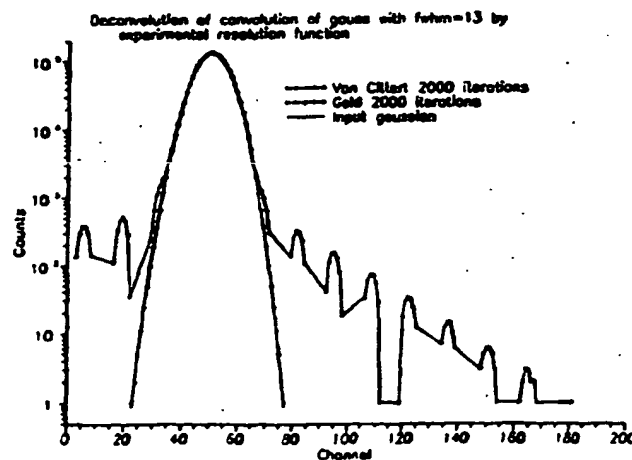


Fig. 3. Comparison of peak deconvoluted by Van Cittert and Gold method.

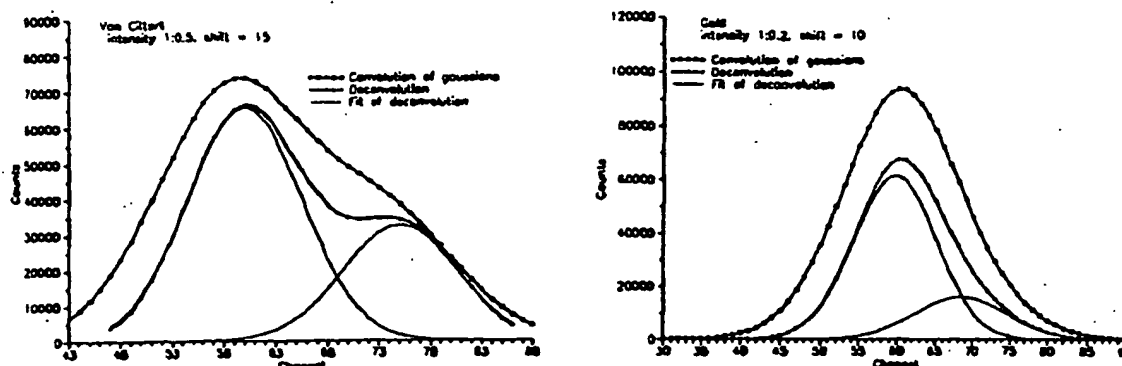


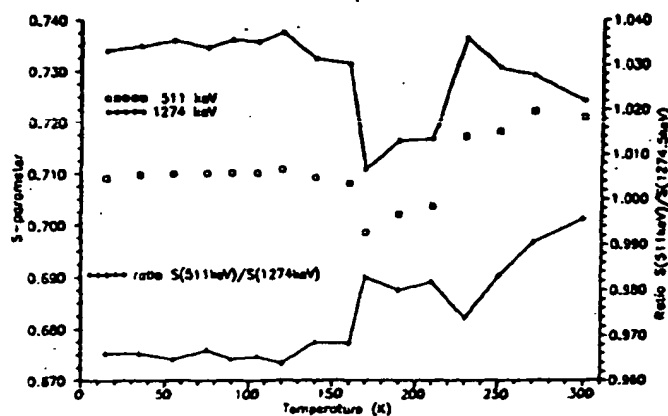
Fig. 4. Convolution and deconvolution of Gaussians.

#### 4. Experimental spectra

All these studies of the convolution of Gaussians and following deconvolution were aimed at finding out the possibility and limitations of the deconvolution methods and their possible use in the deconvolution of experimental  $\gamma$ -ray spectra. Because the methods are quite reliable (especially Gold's method) we have used Gold's deconvolution method for our experimental spectra.

The effect of DBAL is quite smallscale and one needs a very stable measuring system for its measurement. We can get information about its stability from the 1274 keV peak, which is measured simultaneously with the 511 keV annihilation peak if we use  $^{22}\text{Na}$  as a radioactive source.

In our experiment we used an HPGe detector with a resolution of 1.56 keV at 570 keV with a  $^{207}\text{Bi}$  source. The energy calibration of the system was 163 eV/channel. The number of counts at the annihilation peak was  $\approx 630\,000$  and the number of counts at the 1274 keV peak was  $\approx 180\,000$ . In Fig. 5 we can see the  $S$ -parameter with physical information about the momentum distribution of the electrons and the  $S$ -parameter of the 1274 keV peak, which carries information about the stability of the measuring system. If there was no instability during the measurement, it should be a constant. As we can see, the  $S$ -parameter of the 1274 keV peak was not constant, so the measuring system was not stable. So we do not know if the structure in the  $S$ -parameter of 511 keV was really a consequence of this instability or if it had a real physical meaning. Of course, there is a possibility to try to use the  $S$ -parameter of the 1274 keV peak as a correction on the instability, e.g. by calculating the ratio  $R = S(511\text{ keV})/S(1274\text{ keV})$ . The result of such procedure is shown in Fig. 5. As we can see, this simple procedure does not suppress the effect of instability. Because we are

Fig. 5.  $S$ -parameters without the deconvolution procedure and the ratio  $R$  of the  $S$ -parameter at 511 keV to the  $S$ -parameter at 1274 keV.

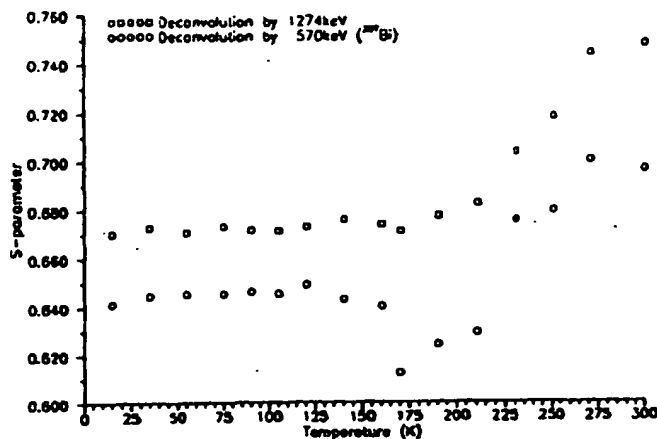


Fig. 6. S-parameters after deconvolution.

interested only in the relative changes of the  $S$ -parameter on temperature, we can make a deconvolution of the 511 keV peak with the 1274 keV peak and try to suppress the effect of instability.

Fig. 6 shows the result of these deconvolutions (the  $S$ -parameter after deconvolution) along with the  $S$ -parameter from deconvolutions of the 511 keV peak with the 570 keV peak ( $^{207}\text{Bi}$ ), which was measured at the beginning of our experiment. So we can see that the structure at the temperature of 175 K was only a consequence of instability of the measuring system, because it disappeared after the deconvolution of the 511 keV peaks with the 1274 keV peaks. We can obtain physical information from the dependence of the  $S$ -parameter on the temperature by using a deconvolution procedure.

This procedure can also be used for a reliable estimate of the electron momentum distribution. In such case we need a better resolution function (e.g. 514 keV from the decay of  $^{85}\text{Sr}$ ) measured simultaneously with the annihilation line.

## 5. Conclusion

It turned out that the described deconvolution methods are in principle convenient for processing experimental spectra from DBAL measurements. They help to suppress influence of instability of the measuring system. Particularly the Gold deconvolution method works very well and it does not deform the deconvolution result. The reproducibility of the deconvolution results tested on simulated convolutions of Gaussians is very good. Van Cittert's algorithm gives the results with oscillations from positive to negative values on the wings of the deconvoluted peaks. Gold's deconvolution gives only small non-negative oscillations. Unambiguously we can say that Gold's algorithm is more reliable with regard to the precision of the deconvolution. The time needed for the deconvolution is practically the same for both algorithms.

So, the use of Gold deconvolution is a cheap and simple way to eliminate the electronic instabilities which blur physical effects.

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